REPORT OF BUREAU OF MINES RESEARCH AND TECHNOLOGIC WORK ON COAL AND RELATED INVESTIGATIONS, 1955

BY E. P. CARMAN, M. R. GEER, AND H. L. RILEY

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INTRODUCTION AND SUMMARY

This report is the 20th in a series summarizing Bureau of Mines research and technologic work on coal and related investigations; it is the second[4] to be issued on a calendar-year basis. Previous reports related largely to publications by Bureau authors during the reporting period. This one discusses the publications issued during 1955 and in addition describes briefly the work in progress during the year. Further details on the published work may be found by referring to the publications cited in the footnotes.

The Bureau continues to direct its research and technologic work on coal toward safer and more efficient mining, improved preparation, and more effective utilization, with the ultimate twin goals of promoting conservation and insuring that coal will make its maximum contribution to the Nation's industrial capacity. Significant advances toward these goals were made in 1955.

Other Government agencies turn to the Bureau for asssistance in the problems involving fuel. They receive consulting services ranging from the preparation of coal specifications to combustion engineering. The coal sampling and analysis program is a part of this service work, for it provides the data that serve as a basis for the entire Government coal-purchasing program. Also related is the work on boiler feedwater, extended to all Government-operated boiler plants. These functions of the Bureau required chemical analyses of nearly 10,000 coal samples and over 13,000 samples of boiler feedwater during 1955. However, this service work increases the efficiency and economy of fuel utilization, saving the Federal Government many times its nominal cost.

In the never-ending search for improved coal-analysis techniques, the losses in heating value during the standard air-drying procedure and during storage of samples in sealed containers were measured. Several analytical methods proposed by the International Organization for Standardization were examined, and cooperation was continued with the Coal Committee of the Economic Commission for Europe in its efforts to establish an international standard system of coal classification.

Ten county reports on the recoverable reserves of coking coal in the Appalachian coal fields, covering counties in Pennsylvania, West Virginia, and Tennessee, were published during the year, bringing the total of such reports to 31. All further studies on reserves will be made by the Federal Geological Survey, but the Bureau will continue to report the preparation and carbonization characteristics of coking coals. In these 2 series reports were issued on the preparation characteristics of coals from 1 county in West Virginia and 2 in Kentucky; the carbonizing properties of coals in West Virginia, Kentucky, Tennessee, and Pennsylvania were reported.

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[4] Carman, E. P., and Caldwell, D. C., Report of Research and Technologic Work on Coal and Related Investigations, 1954: Bureau of Mines Inf. Circ. 7756, 1956, 80 pp.

In Alaska drilling has been recessed in favor of trenching as a means of obtaining geological information pertinent to the search for additional coal reserves; a prime objective of this program is to insure that Alaska will not depend on waterborne fuel in a national emergency.

Investigations intended to develop improved mining methods were carried forward in both anthracite- and bituminous-coal mines. In the former the use of collapsible steel roof supports was extended, and further operating data were obtained on the Bureau-designed vibrating-blade coal planer. In bituminous-coal mines study was continued on application of the German coal planer to longwall mining of thin beds. Also, an investigation was undertaken of the influence of bed thickness on mining methods, types of mechanical equipment, and rate of progress in both development and production operations. The use of auger mining to recover coal from the highwalls of strip pits was the subject of another study.

The Bureau of Mines has been engaged in five foreign coal projects sponsored by the Interntional Cooperation Administration, United States Department of State. As part of the United States Government's technical assistance to underdeveloped countries, the Bureau's efforts have been directed toward training foreign engineers in the United States and abroad. Programs in Afghanistan, Colombia, India, and the Philippines were instigated to increase the local supply of solid fuels. A Bureau engineer has been advising the Government of Pakistan on engineering details involved in constructing an ammonium sulfate fertilizer plant in that country.

Both pilot-plant investigations and tests of full-scale equipment demonstrated the feasibility of conserving fuel resources by recovering usable coal from washery refuse, as well as from beds formerly considered too impure to work. Means of increasing the capacity of the kerosine flotation process for cleaning fine coal were reported. The results of washability studies and cleaning tests on coals from Arizona and Montana were published; similar work was done on Alaska coal and Pennsylvania anthracite.

Surveys of lignite preparation and burning practices were conducted as part of the Bureau's investigations concerned with utilization of low-rank coals. Experimental work disclosed that lignites from the North Central States had Hardgrove grindability indexes ranging from 26 to 87, varying with moisture content of the lignite.

Research in the field of heat transfer and fluid mechanics was directed toward improving the efficiency with which coal of all ranks can be burned.

One means of extending our reserves of metallurgical coking coal is substitution of other fuels that are in ample supply. Therefore the use of anthracite in foundry cupolas, blast furnaces, blast cupolas, and the electric furnace was investigated. Substituting up to 35 percent of anthracite for coke in a blast cupola was successful.

Coal-carbonization studies included the effects of process variables on product yields, the expansion of coal during carbonization, and the influence of various pretreatments on coking. Low-temperature carbonization studies of western coals were continued, and an investigation of the relation between petrographic and carbonization properties of western coking coals was initiated with the cooperation of industry. Low-temperature-tar research was accelerated at the Lignite Experiment Station, Grand Forks, N. Dak., and a new laboratory for this work was established at the Appalachian Experiment Station, Morgantown, W. Va.

Nickel was shown to be superior to iron as a catalyst in the methanation of synthesis gas to form a high-B.t.u. gas. Further tests in the area that had been hydraulically fractured at the underground-gasification project, Gorgas, Ala., showed that, to preserve bed permeability, it was necessary to ignite the coal at the gas outlet, so that the ignition zone would travel toward the air inlet.

Investigations were continued on the gas-synthesis and coal-hydrogenation processes for converting coal to synthetic fuels to obtain both fundamental and engineering data. The structure of coal has been studied to obtain information on its behavior in chemical processes. Analytical methods were developed for process control and product characterization.

Pilot-scale coal-gasification tests at atmospheric pressure, using oxygen and steam with various ranks of coal, showed that the percentage of carbon gasified increased with decrease in the rank of coal for given carbon-input rates and reactant ratios. Methods studied for feeding coal to pressure gasifiers included use of a slurry under pressure and feeding fine coal into high-pressure, superheated steam. Gas-purification studies were made on various methods of removing dusts, carbon dioxide, hydrogen sulfide, and organic sulfur compounds from synthesis gas. Problems studied relating to the use of nuclear energy to supply the heat required for coal gasification included costs, materials of construction, and operating conditions. Other coal-gasification-research problems investigated at laboratory scale included a kinetic study of the steam-carbon reaction, flashing and gasification of coal slurries, and the use of metal chelates as oxygen carriers.

Research on the use of iron catalysts in the form of lathe turnings showed that a low resistance to gas flow was obtained with such catalysts, thus making the hot-gas recycle process more feasible. Nitrided, fused-iron catalysts were employed successfully in both the oil-circulation and fluidized reactors. Large yields of alcohols and other oxygenated molecules were produced by both processes. Laboratory investigations have been made on problems of suspension or settling of catalyst particles in slurry operations. Equilibrium studies on the absorption of hydrogen sulfide in potassium carbonate-bicarbonate solutions indicate that rather complete removal of hydrogen sulfide in the presence of carbon dioxide can be achieved in the hot-carbonate scrubbing process, provided that highly regenerated carbonate solutions are employed.

Small-scale studies were continued on the development of improved fused and massive iron catalysts. The sulfur poisoning of iron catalysts is being studied to determine the practical limits of sulfur concentration that can be tolerated in the feed gas and to obtain basic data regarding the poisoning of the catalyst and possible methods of regeneration. A gas chromatographic method has been developed to determine the extent of branching in C_4 hydrocarbons. Electron-microscopy studies of carbon deposited from carbon monoxide on iron, cobalt, and nickel show that carbon is deposited in the form of filaments containing nuclei of a metal or a metal carbide. Anomalies in the Curie point of Hägg iron carbide have been investigated.

Studies of the chemistry of carbon monoxide and carbonyls were continued, and data are reported on the effect of the structure of olefinic hydrocarbons on the rate of hydroformylation reactions in the presence of cobalt carbonyl. Special analytical methods have been developed for characterizing the products from the Fischer-Tropsch synthesis.

Bench-scale coal-hydrogenation studies were made at elevated temperatures (500°-525° C.) to determine the operating characteristics of a small, tubular reactor for converting coal into gasoline in a single step. Also, pilot-plant experiments on the high-temperature hydrogenation of coal have been made on Rock Springs, Wyo., coal, impregnated with ammonia molybdate, using a topped, high-temperature tar as a vehicle. Depending upon the amount of catalyst used, temperature control and coke formation gave difficulties.

Hydrocarbonization studies (that is, carbonization of coal in hydrogen at moderate pressures) were made in the small, semicontinuous hydrogenation unit to develop means of avoiding agglomeration of coal during this treatment. It was found that agglomeration tendencies depended on the rank and size of coal and the method of heating.

Coal-hydrogenation experiments and tests in batch autoclaves have shown variations in the activity of selected catalysts at certain pressures and temperatures, indicating that relative activity at a particular pressure cannot always be predicted from knowledge of activity at a different pressure.

In studies on the structure of coal, a program of microbial degradation of carbonaceous material was initiated. Bacteriostatic and fungistatic materials have been extracted from coal, and their potency has been determined. Microflora associated with lignites have been isolated and identified. Also, the similarities and differences among the infrared spectra of coal and related substances have been determined. Methods of X-ray-diffraction analysis are being developed to evaluate the parameters of coal. Provisionally, the characteristics of Bruceton anthraxylon were determined. Sorption studies on coal, using normal butane and isobutane, indicated that a large fraction of the pores of coal have openings of about 5 Å. units. Several coal-hydrogenation oils and low-temperature lignite tar have been found to contain optically active constituents. Fractions isolated from these sources give both positive and negative rotations.

Methods have been developed for identifying phenols by X-ray diffraction and characterizing aromatic mixtures by ultraviolet spectrometry.

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This report describes work performed under the technical direction of many members of the staff of the Bureau of Mines, who cooperated and assisted in the preparation of data and material for use in this report. A number of utilities and power plants in the Minnesota-North Dakota area assisted in a study of the use of lignite for power production by giving data on their operations. Many coal and coke operators in the Appalachian region, too numerous to mention individually, have cooperated with the Bureau of Mines in various ways, such as providing or allowing the collection of samples, permitting inspection of their facilities, or furnishing requested data. During the period covered by this report, the following companies, institutions, and Federal and municipal agencies have made financial, research, or technological contributions to the Bureau of Mines program. Their cooperation is hereby gratefully acknowledged.

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Amherst Coal Co.
American Society of Mechanical Engineers.
Amos, A. R., Co.
Atomic Energy Commission.
Baltimore, Md., City Council.
Boeing Airplane Co.
Bureau of Reclamation, U. S. Department of the Interior.
Cambria Clearfield Mining Co.
Carborundum Co.
Cardox Corp.

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Central Electric Power Cooperative, Inc., Minot, N. Dak.
Christopher Coal Co.
Clearfield Bituminous Coal Corp.
Coaldale Mining Co.
Colorado Fuel & Iron Corp.
Colorado School of Mines Research Foundation.
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Esso Research & Engineering Co.
Excavators, Inc.
Ford & Gaskill Coal Co.
Ford Motor Co.
General American Transportation Corp.
Glen Alden Corp.
Grafton Coal Co.
Gulf Smokeless Coal Co.
Halliburton Oil Well Cementing Co.
Hanna, M. A., Co., Susquehanna Collieries Division.
Hudson Coal Co.
International Cooperation Administration, U. S. Department of State.
Island Creek Coal Co.
Jeddo-Highland Coal Co.
Johnstown Coal & Coke Co.
Kaiser Steel Corp.
Kemmerer Coal Co.
Kentucky Department of Mines and Minerals.
Koppers Co., Inc.
Lehigh Valley Coal Co.
Moffat Coal Co.
Norton Co.
Pennsylvania Coal Co.
Pennsylvania Crusher Co.
Pine Township Coal Co.
Pittsburgh Coal Co.
Princess Coal Sales Co.
Reading Anthracite Co.
Red Jacket Coal Corp.
Rochester & Pittsburgh Coal Co.
Standard Oil Development Co.
Tennessee Coal & Iron Division, U. S. Steel Corp.
Tennessee Valley Authority.
Texas Power & Light Co.
Union Carbide & Carbon Co.
Union Pacific Coal Co.
University of Alabama.
University of Maryland.
University of North Dakota.
University of Washington.
University of West Virginia.
U. S. Pipe & Foundry Co.
U. S. Public Health Service
U. S. Steel Corp.
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Virginia Department of Labor and Industry.
Warner Collieries Co.
Westinghouse Electric Corp., Atomic Power Division.
Weston Dodson Co.
White, J. D., Engineering Co.
Youghiogheny & Ohio Coal Co.

ORIGIN, COMPOSITION, AND PROPERTIES OF COAL AND RELATED MATERIALS

Inspection, Sampling, and Analysis

The fuel-inspection and analysis service maintained by the Bureau serves as the basis of the Government's fuel purchasing on specification and bids; however, the coal analyses accumulated in the course of this work are made available to the public and are widely used in private purchasing of industrial coal. Government purchasing officers are advised on technical matters relating to coal, assisted in determining awards of contracts, and aided in carrying out contract terms.

The sampling and analysis service has achieved substantial savings to the Government by making it possible to award coal-purchase contracts on a guaranteed-quality basis and to adjust prices when shipments fall below contract specifications. It also provides valuable data for use in evaluating future bids.

During the year coal was produced equal to or in excess of demand, and Federal agencies had no difficulty in obtaining bids covering their requirements. All agencies except the Post Office Department purchased on a guaranteed-analysis basis, including penalties for delivery of substandard coal.

Coal requirements of Federal agencies for the period, except the Atomic Energy Commission, Tennessee Valley Authority, and coal purchased for export, were estimated at approximately 5 million tons. Requests from Federal agencies for analyses for use in evaluating coal bids continued at about the same rate as in the previous year because of the large number of bids and strict adherence to specification requirements regarding the elimination of mines on which there were no Bureau of Mines analysis records. Contracting for coal for the fiscal year 1956 began in February and continued through June. Recommendations were made for awards of contracts for some agencies.

Proximate or ultimate, sulfur, and calorific-value analyses were reported on 9,569 samples; ash-softening-temperature determinations were reported on 2,386 samples; free-swelling indexes were computed on 368 samples; Hardgrove grindability was determined on 237 samples from purchases of coal; and tipple and breaker inspections were made in connection with Federal coal purchases. Analyses were reported on 934 tipple samples collected at 321 mines in 17 States and Alaska and on 542 samples of export coal, representing approximately 910,000 gross tons shipped overseas by the FOA.

Arrangements were made with the Atomic Energy Commission to supervise and direct the sampling and analysis of coals delivered to the Savannah River, S. C., and Dana, Ind., plants and to provide the services of technical and other personnel to conduct the work; operations were started at Savannah River in October and at Dana in December.

In Alaska the Bureau's coal-analysis laboratory was moved to new, more suitable quarters at Elmendorf Air Force Base. Some 70 analyses were made there, representing 63,000 tons of coal delivered to military posts in the Anchorage area. In future, samples from all the principal military installations in the Territory will be analyzed here, speeding settlement of coal contracts. Samples originating from the Bureau's exploration program also are analyzed, and military personnel is trained in sampling.

During 1955 the coal-analysis laboratory at Pittsburgh, Pa., analyzed 37,134 samples of coal, coke, char, tar, coal-mine dust, and related material. This number included 23,055 samples from the Branch of Coal-Mine Inspection, collected during 4,277 inspections in 22 States and Alaska to carry out the provisions of Public Law 552, Federal Coal Mine Safety Act; 9,907 represented Government purchases of coal on a penalty basis and tipple inspections (56 percent of this group came from branches of the United States Department of Defense); and 4,172 were submitted by Bureau of Mines and other Government organizations in connection with research and exploration projects on coal. Analysis of the 37,134 samples required 363,688 chemical and physical tests.

Examination of Coal-Analysis Methods

Present methods used in analyzing coals and coal-mine dusts are being studied constantly to adapt modern improvements to analytical apparatus and laboratory techniques. New methods are investigated and old ones critically examined to reduce time and operating costs and to increase accuracy.

Tests were continued during the year on the loss in heating value of six coal samples while in storage. It was determined that all ranks of coal lose some heating value when stored in sealed containers. The losses for 1 month of sealed storage ranged from 0.5 percent for a high-rank bituminous coal to 1.5 percent for a low-rank coal; the losses increase with storage time. Additional losses in heating value occur during the normal air-drying of samples before preparation for analysis. Such losses amount to about 0.1 percent for a high-rank bituminous coal and 0.5 percent for a high-moisture, low-rank coal.

Single- and a double-crucible methods were proposed by the International Organization for Standardization for international use in determining volatile matter in coals. Both methods were applied to different ranks of American coals and the results compared to values obtained by ASTM methods. The two proposed methods gave values comparable to those obtained by ASTM methods; however, the double-crucible method is very time-consuming and not considered practical for routine analysis. In the single-crucible method, a cylindrical silica crucible is placed on a refractory base and inserted into a muffle furnace at 925° C. for 7 minutes. It is weighed after cooling in a desiccator and the volatile matter determined from the loss of weight.

A new rapid method was developed for evaluating coal-mine dusts for incombustible content. The method employs a modified volumeter to obtain the specific volume of coal-mine dusts. The specific volumes of hundreds of samples were correlated with incombustible contents obtained by chemical methods, and a conversion table was prepared for evaluating unknown samples. A few samples gave an indicated incombustible content that differed by over 5 percent from the value obtained by chemical analysis; the method is not considered accurate enough to replace the chemical method for all samples. However, it has been recommended to the Health and Safety Activity to be used in estimating the incombustible content of a large percentage of the samples, thereby reducing the workload in the laboratory for the chemical method of analysis.

Low-Temperature-Carbonization Assay

A Bureau of Mines bulletin has been nearly completed covering results of Fischer-Schrader low-temperature-carbonization assays of about 400 coals from nearly every coal-producing State, Alaska, and British Columbia, Canada. The coals tested cover a range of rank from lignite to semianthracite. The report lists the yields of carbonization products and includes analyses of the gas, the types of carbonized residue obtained, and chemical analyses of the coals. Also included is a description of the procedure used in making the assay, details of which have not been published previously.

A carbonization-assay survey of lignites by the Lignite Research Laboratory was completed during the year. Initiated to delineate bench-scale carbonization characteristics of lignite samples obtained from various localities in the North Central area and, insofar as possible, geologically different beds, the survey has included face samples from 23 mines of varying size and 10 drillhole cores available to the laboratory.

Assays conducted during 1955 include samples of Fleissner-dried lignite produced at this station and cores obtained in drilling by the Great Northern Railway. A short series of comparative tests conducted with the Denver laboratories on identical blends of coal indicated good reproducibility of the modified Fisher assay between different laboratories.

Yields of products from selected samples to indicate extremes showed char recoveries on the moisture- and ash-free basis ranging from 56.7 percent for slack lignite or "leonardite" to 80.8 percent for a concentrate of the fusain component from a typical face sample; tar plus light-oil yields ranged from 3.3 percent for the fusain concentrate to 11.0 percent for a normal sample; gas recoveries varied more widely both in yield and in composition, ranging from 10.0 to 27.7 percent, the latter being from leonardite and quite high in carbon dioxide. Overall results of the survey indicate a general uniformity of north central United States lignite with respect to low-temperature carbonization. The humic lignite of the area produces less tar and light oil than does the xyloidal lignite of other localities, averaging approximately 8 percent, based on moisture- and ash-free lignite. Recoveries of char average approximately 70 percent on the same basis. Although the survey failed to demonstrate significant variation in lignite from different localities or different beds, it does provide a basis for estimating recoveries to be expected on large-scale, low-temperature carbonization.

Analysis of Miscellaneous Materials

Total silica was determined in 87 samples of rock dusts submitted by Federal coal-mine inspectors to determine compliance with Public Law 552, Federal Coal Mine Safety Act, which specifies that rock dust used in coal mines shall contain not more than 5 percent total silica. Samples of eight other rock dusts were examined to determine their suitability for rock-dusting coal mines; the examination consisted of tests for size and caking properties and determination of the total silica content.

In all, 189 samples of various materials analyzed or tested in connection with Bureau investigations of preparation and utilization of coal and of accident prevention included: 133 coals and pitch-coke mixtures examined for estimation of coking properties by the agglutinating-value test; 19 coal and coke ashes; 3 manganese-ore dusts; 5 coal cores for determination of oil content; 3 layer samples of coal from the Chilton coal bed for low-temperature carbonization assays and agglutinating-value tests; 6 samples consisting of an enrichment of petrographic components separated from Chilton-bed coal for Audibert-Arnu dilatometer tests; 4 coke samples for residual tar; and 16 miscellaneous samples.

Analyses of Ash

A report is being prepared that lists the chemical composition of 323 samples of coal ash from 24 States and Alaska. Composition of coal ash as it relates to the flow properties of slag is important in connection with the design and operation of slag-tap pulverized-coal furnaces, cyclone furnaces, and equipment for complete gasification of pulverized coal to make synthesis gas. The composition of coal ash also is important in selecting coals for special purposes, such as the manufacture of cement and the burning of the finer grades of ceramic products. In addition to the ash analyses, the report includes a discussion of the nature and occurrence of ash-forming mineral matter in coal and the relationship of ash composition to fusibility.

Classification, Constitution, and Petrography

International Classification and Analysis of Coal

Increased world trade in coal since World War II necessitated development of international methods of analyzing, testing, and classifying coals to provide a common language for evaluating coals of various countries. The Coal Committee of the Economic Commission for Europe established a Classification Working Party in 1949 to undertake development of an international classification system for coal. Also, Technical Committee 27 on Solid Mineral Fuels, International Organization for Standardization (ISO), in cooperation with standardization bodies of various countries has undertaken work on international standardization of methods for analysis and testing of coal. At present, active cooperation in the work of both committees is being given by nearly every European country, the United Kingdom, and the United States. A classification system for brown coals and lignites is now being formulated. Other work underway includes schemes for classifying coal by size and for classifying coke.

In connection with the classification of low-rank coals, exchange samples of a subbituminous B coal from Alberta, Canada, and a subbituminous A coal from Selangor, Malaya, were analyzed and tested to supplement similar analyses made during the previous year on low-rank coals from other countries. Information also was supplied to the Secretariat of the Coal Committee regarding methods used in the United States for analysis and testing of coke and Federal specifications used by Government agencies for purchasing foundry and domestic coke.

In the work on developing a scheme for coke classification, delegates were requested to submit comments regarding a proposed tentative scheme under consideration by the Classification Working Party. This tentative scheme was submitted to a selected group of coke technologists in the United States for study and comment. As far as American conditions are concerned, many replies questioned the need or usefulness of the proposed system on classifying coke. Nearly all stressed the need of standardizing test methods for evaluating coke for metallurgical use.

At the fourth meeting of ISO Technical Committee 27 held in Stockholm, Sweden, June 6-10, 1955, items considered were methods for determining moisture, ash, volatile matter, carbon, hydrogen, sulfur, forms of sulfur; swelling and coking tests; phosphorus, chlorine, arsenic, tar yields from brown coals and lignites; and mineral matter.

In cooperation with ISO Technical Committee 27 in developing and evaluating methods for analyzing and testing coal and coke, two laboratory investigations were made during the year. One was an investigation of methods of determining moisture; the other was a preliminary study of a method for the direct determination of mineral matter in high-rank coals.

ISO tentative recommendations for determining moisture now specify that such determinations be made in an atmosphere of nitrogen. To determine whether air could be substituted for the nitrogen atmosphere, various methods of determining moisture in 19 typical American coals of representative ranks were investigated. Methods were: (1) ASTM oven method, using dry air; (2) indirect method in oven, using dry, oxygen-free nitrogen; (3) direct method, using dry, oxygen-free nitrogen; and (4) azeotropic distillation with toluene.

In general, the difference between individual determinations showed that the precision of the moisture determination is good for all methods investigated. For all but the azeotropic distillation method, different heating periods were used to determine the effect of time of heating. One hour was enough for the ASTM method. Except for the 2 lignites and 1 subbituminous C coal, 1 hour of heating was satisfactory for the indirect oven method in nitrogen. For these 3 low-rank coals, 2 hours was necessary to reach constant weight by this method. One-half hour of heating by the direct gravimetric method was adequate for all of the coals but the two lignites. For lignite and subbituminous coals, more tests are to be made to determine the heating periods required to reach constant weight.

Except for the two lignites, the ASTM air-oven method gave slightly lower results than the ISO direct gravimetric method, indicating some oxidation of the sample when heated in air. The loss-of-weight-in-nitrogen method checked closely with the direct gravimetric method, except for the two lignites and the subbituminous C coal. In general, the azeotropic distillation method gave slightly higher results than the direct gravimetric method. For the higher rank coals, the differences were minor.

It was concluded that, because the conventional methods of heating at 105° to 110° C. are empirical in nature and all of the moisture is not removed at these temperatures, such refinements as heating in nitrogen are of questionable value in analyzing coal for commercial purposes. A report covering the investigation was submitted to ISO Technical Committee 27 on Solid Mineral Fuels.

A method for the direct determination of mineral matter in coal was applied to three coal samples from Pennsylvania, Iowa, and Kansas. The method is based on elimination of mineral matter by treating the coal with hydrofluoric and hydrochloric acid, according to a method proposed by a German scientist. The average results of direct determination of mineral matter in coals from Pennsylvania, Iowa, and Kansas were 7.81, 18.69, and 16.32 percent, respectively. Duplicate determinations checked satisfactorily. For comparison, the mineral-matter contents were computed from the analyses, according to a formula developed by British scientists and known as the King, Maries, and Crossley formula. The calculated values for the 3 coals were 7.51, 18.11, and 15.22 percent, respectively. These tests indicate that the direct method has considerable merit and should be studied further on additional coals.

Coal Petrography and Paleontology

Since 1930 the Bureau of Mines has carried on systematic studies of the physical composition of American coals by petrographic methods as a contribution to fundamental knowledge. Representative vertical sections, called column and core samples, of various coal beds have been collected from mines and diamond-drill test holes for making thin, translucent slices, by means of which qualitative and quantitative microscopic determinations were made of the plantlike ingredients of which all coal is composed. Such microscopic analysis provides data for systematically grouping the different varieties of coal into physical types defined in accordance with the Bureau's system of coal description and classification. This information, pertaining to 217 American coals, has been assembled into a Bureau report to be published soon.

A series of investigations was begun on the relation between the microscopic composition and the chemical and physical properties of banded bituminous coals. The physically heterogeneous, high-volatile A coal of the Chilton bed in Logan County, W. Va., was the source of column, face, and channel samples obtained for the

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initial study. Preliminary work included a microscopic and type classification of coal of the column sample. This analysis revealed that the petrographic types bright, semisplint, and splint coals made up the bulk of the coal. The bright coal contained an unusually rich concentration of anthraxylon, and the semisplint consisted largely of spores, whereas the most conspicuous microconstituent of the splint was opaque attritus. Samples of the three coal types were cut from typical petrographic layers for various analyses and tests.

A manuscript is in preparation, intended as a report of investigations, describing the results of a detailed study of variation in swelling properties of the petrographic constituents of the Chilton coal. Ninety-four incremental samples, each representing about 3/4 inch of the column, were subjected to free-swelling tests. These tests showed a wide range of swelling numbers from 1 to 8, with an average of 5-1/2 for whole coal. As a rule, bright coal was strongly swelling, ranging from index number 4 to 8. The splint-coal samples were weakly swelling or nonswelling. Anthraxylon thus proved to be the principal substance of the Chilton coal responsible for its swelling capacity. Fusain was determined to be totally inert. The tests indicated that opaque attritus and mineral matter also were inert. Translucent humic matter did not appear to have the property of swelling appreciably but did become plastic when heated and acted as an agglomerating agent.

Anthraxylon, translucent humic matter, and opaque attritus are the three most abundant microconstituents of the Chilton coal. Anthraxylon usually predominates in bright coal, translucent humic matter in semisplint and cannel coals, and opaque attritus in splint. Therefore, the swelling property of any particular sample seemed to be a function of the proportional amounts of these three substances in the coal. The ingredients present in minor quantities, such as spores, resin, fusain, and mineral matter, are usually unimportant to swelling but may have a cumulative effect; in some instances, one or more of these may be concentrated in the coal and have a marked effect on its swelling.

Because of the speed with which preparations of fossil pollen from a Tertiary coal can be made, paleontology is a most rewarding technique in terms of yield of information about the flora that contributed to coal. In view of this, a study of pollen from a small deposit of lignite or brown coal in the town of Brandon, Vt., though made in large part at Harvard University, was published by the Bureau.5/ This study revealed that several groups of plants present in the Brandon lignite were those found in wet, tropical, or subtropical climate. A significant discovery in this study was a group of which the most nearly equivalent plants now living are found in the bays and swamps of the southeastern United States.

Microreflectivity Analysis of Coal

Petrographic analysis of coal has proved valuable, particularly in Europe, in controlling blending practices for making metallurgical coke and has been shown to be applicable to the prediction of yields of liquid products in the hydrogenation of coal to synthetic oils and gasoline. However, present methods of making such microscopic studies of coal are largely subjective, and a need has been expressed for more objective analytical procedures that may have even greater value in controlling utilization processes.

Reflectivity is a physical property that is basically involved in the microscopic differentiation of various coal components, but present analytical methods

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5/ Traverse, Alfred, Pollen Analysis of the Brandon Lignite of Vermont: Bureau of Mines Rept. of Investigations 5151, 1955, 107 pp.

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make little use of quantitative determinations of this property. Probably the major reasons for neglecting this objective measurement in routine petrographic analysis lie in the relative difficulty of measurement and the excessive time required in manual methods of operation. To attempt to overcome these objections, the possibility of using a scanning device and a photoelectric detector to measure reflectivity variations in coal samples has been studied, and the results have been reported.[6]

Graphs showing the proportions of components of different reflectivity in a sample are obtained by scanning the specimen and recording, by means of a highly sensitive photomultiplier photometer, the intensity of light reflected through a fine aperture that permits high resolution of minute areas of different reflectivity. The tests can be made on either solid blocks of coal or on granular samples molded into briquets with a binder. This preliminary study has been made with a relatively slow recording microammeter, and analysis of the chart record is rather laborious, but it would be quite feasible to use electronic circuits for recording and totalizing the results in a much shorter period.

Probably the most useful information readily obtainable from the reflectivity-distribution curves of various coal samples is the fraction of the coal that has reflectivity greater than that of the main anthraxylon component. Such material will be relatively inert compared with the anthraxylon, it will liquefy less readily when hydrogenated, and it will tend to weaken the coke product if too large amounts are present. Knowledge of the proportional amount of these higher reflective components and a measure of their relative inertness as presented by their degree of reflectivity can thus be gained by a rapid test based on objective measurement of a definite physical property.

Removal of Moisture From Lignite in Inert Atmospheres

Using nitrogen and helium as inert gases, results of experiments indicate that the release of water and carbon dioxide from lignite is a continuous and gradual process at elevated temperatures in inert-gas atmospheres. This continued release, plus the effect of varying the gas atmospheres, points to control of rate by a diffusion mechanism - apparently that of diffusion from the surface of release out through the capillary or pore system of the lignite. Attempts to reach a limiting value for or complete recovery of the contained moisture within a reasonable time apparently result in breakdown of the organic material.

COAL MINING

Current research and technologic work on coal mining falls into four broad categories, each having its own specific objectives: (1) The safety activities are charged with the responsibility of finding and eliminating causes of personal injury and hazards to health; (2) studies of mining methods are intended to develop improved practices that will increase labor productivity and thus provide cheaper coal, at the same time minimizing losses of coal in mining; (3) reserve studies provide the factual data needed in guiding the wise use of our coal resources; and (4) technologic assistance to other coal-producing countries is part of the Government's broad program of foreign aid.

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[6] McCartney, J. T., and Hofer, L. J. E., Microreflectivity Analysis of Coal: Anal. Chem., vol. 27, 1955, pp. 1320-1325.

Study of Known Recoverable Reserves of Coking Coal

The Bureau's program of estimating reserves of metallurgical-grade coking coal, which began in 1948, was terminated this year, when these studies were transferred to the Federal Geological Survey. Thirty-one reports, by individual counties, have been published, making available to interested public agencies and the coal industry a detailed appraisal of the coking-coal resources available for metallurgical use in these counties. Reports published in 1955 cover 3 counties in Pennsylvania, 2 in West Virginia, and 5 in Tennessee. The status of 13 additional reports in various stages of completion is indicated by the maps shown in figures 1 and 2.

Washington County, Pa.

The Pittsburgh bed is the most important in Washington County from the standpoint of both present production and recoverable reserves.[7]

Reserves in 5 coal beds, as of January 1, 1954, are estimated to be 4,181 million tons, of which 4,132 million tons is in beds 28 inches and more thick and thus considered recoverable. The average recovery for all beds in this county is 55.3 percent; and, using this factor, the recoverable reserves are estimated to be 2,284 million tons. Maps of the Washington, Waynesburg, Redstone, Pittsburgh, and Upper Freeport beds are included in the report.

Washington County coals are high-volatile A bituminous in rank and because of their contracting properties during carbonization must be blended with low- or medium-volatile coals to improve the physical properties of the coke.

Greene County, Pa.

The Pittsburgh bed is the most important bed in the county from the standpoints of present production and recoverable reserves.[8]

Estimates of reserves were made for 3 beds, in which 6,011 million tons of reserve coal remained on January 1, 1954. There was 5,967 million tons in beds 28 inches and more thick, and, based on an average recovery of 59.5 percent for the county, the recoverable reserves are estimated to be 3,553 million tons. Maps of the Waynesburg, Sewickley, and Pittsburgh beds are given in the report.

All coals in the county are high-volatile A bituminous in rank, and although the Pittsburgh-bed coal cokes strongly, blending with higher rank coals improves the physical properties of coke made from this coal.

Clearfield County, Pa.

The Lower Freeport and the Lower Kittanning beds are the most important in the county from a production standpoint, and the Lower Kittanning has the largest estimated recoverable reserves.[9]

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[7] Wallace, J. J., Dowd, J. J., Provost, J. M., Abernethy, R. F., and Reynolds, D. A., Estimate of Known Recoverable Reserves of Coking Coal in Washington County, Pa.: Bureau of Mines Rept. of Investigations 5109, 1955, 23 pp.

[8] Wallace, J. J., Dowd, J. J., Provost, J. M., Abernethy, R. F., and Reynolds, D. A., Estimate of Known Recoverable Reserves of Coking Coal in Greene County, Pa.: Bureau of Mines Rept. of Investigations 5143, 1955, 22 pp.

[9] Blaylock, D. W., Dowd, J. J., Abernethy, R. F., and Reynolds, D. A., Estimate of Known Recoverable Reserves of Coking Coal in Clearfield County, Pa.: Bureau of Mines Rept. of Investigations 5166, 1955, 36 pp.

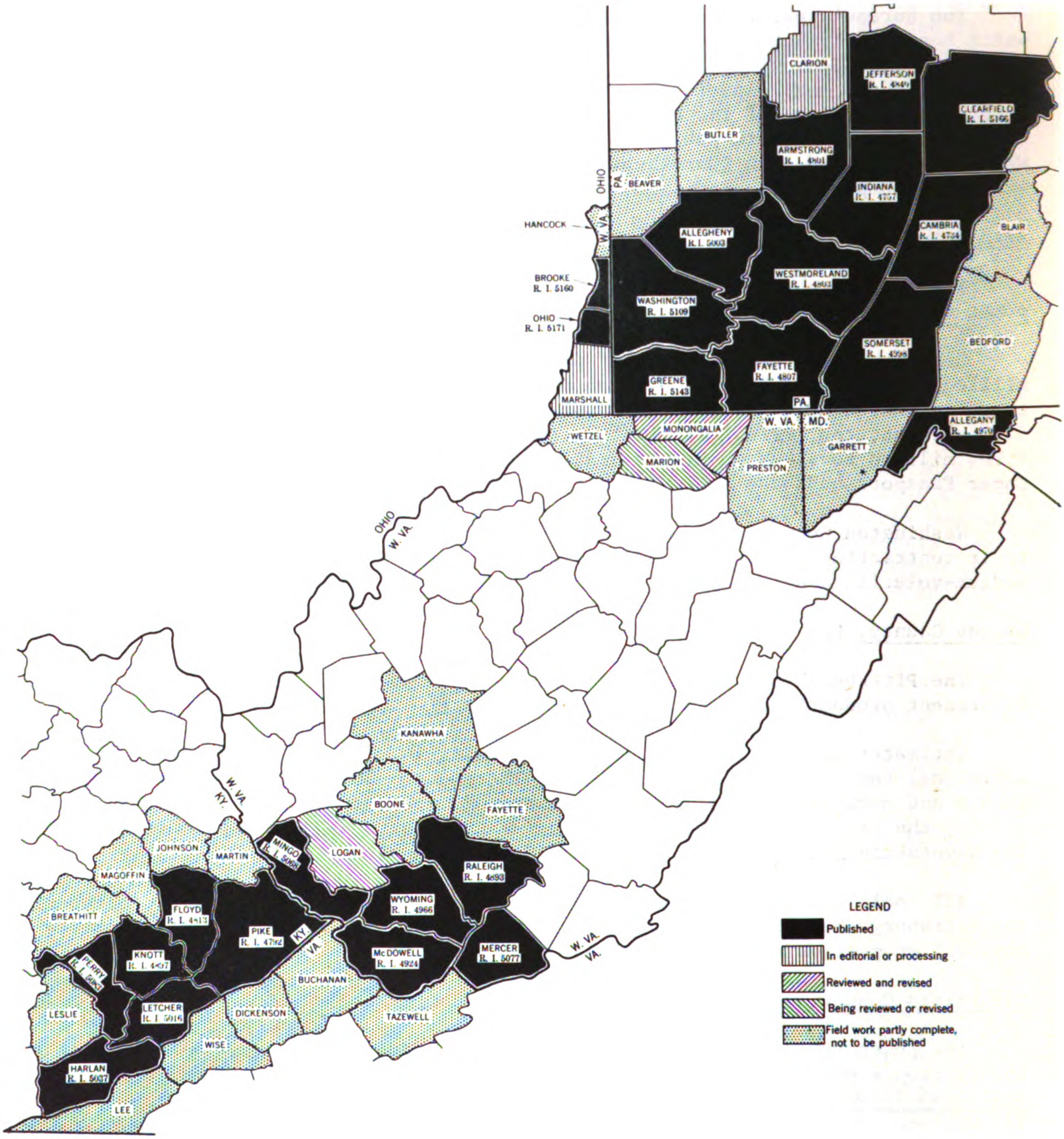

Figure 1. - Status of coking-coal study in Pennsylvania, Maryland, West Virginia, and Kentucky.

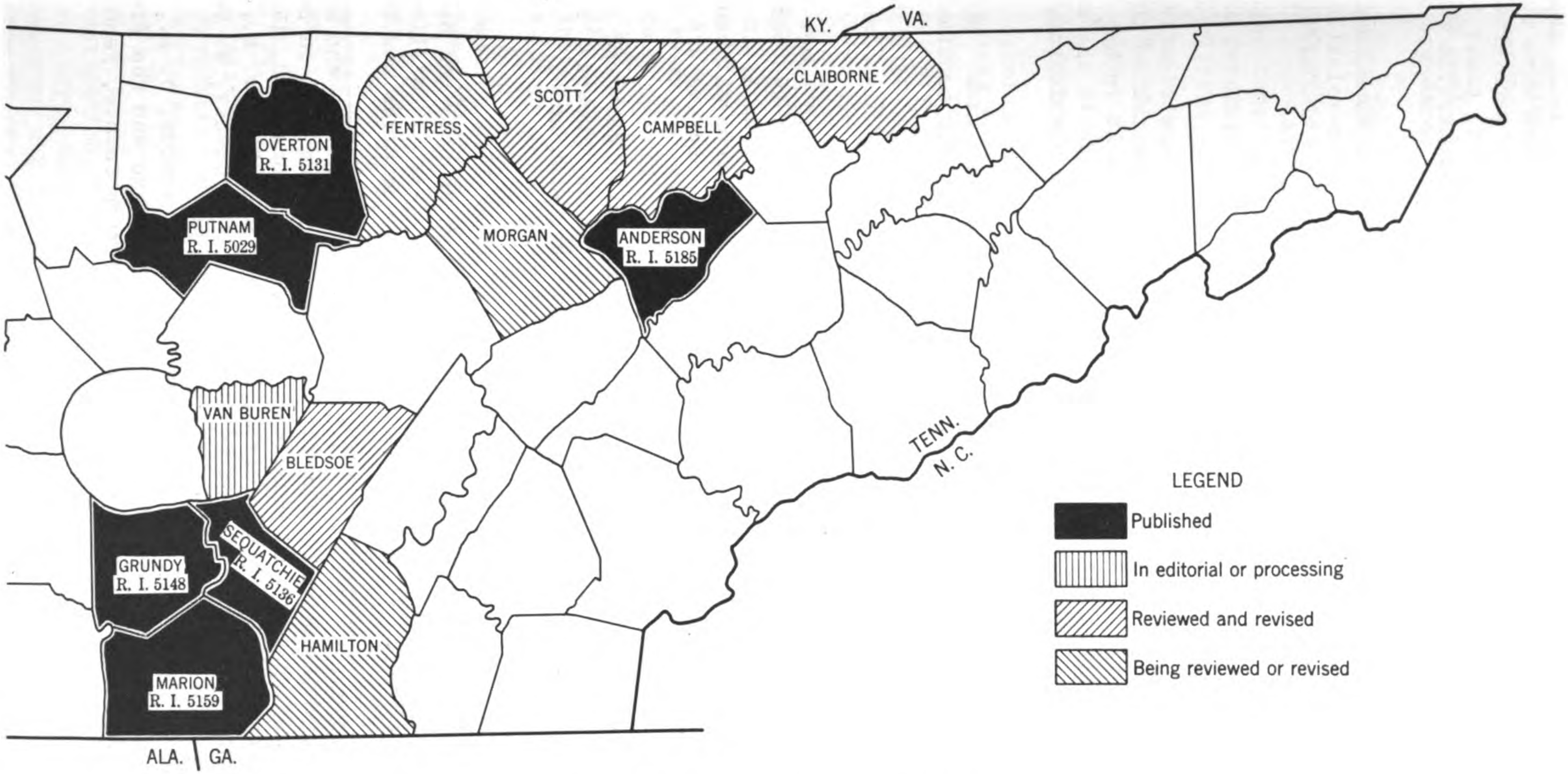

Figure 2. - Status of coking coal study in Tennessee.

Reserves were estimated for 6 coal beds; as of January 1, 1953, the total was 1,909 million tons, of which 1,177 tons is in beds 28 inches and more thick. Based on an average 53.7-percent recovery of coal from all beds, the recoverable reserves are estimated at 633 million tons. Maps for the Upper Freeport, Lower Freeport, Upper Kittanning, Middle Kittanning, Lower Kittanning, and Brookville beds are given in the report.

Clearfield County coals are of medium-volatile bituminous rank, except for a small amount in the northwest corner of the county that is high-volatile A bituminous. Coals from all beds in the county with the exception of the middle Kittanning and Brookfield have been used for making metallurgical coke.

Brooke County, W. Va.

The Pittsburgh bed is the most important in the county, contributing nearly the entire production of coal.[10/] Reserves in 3 coal beds were estimated to be 191 million tons, as of January 1, 1954; 190 million tons is in beds 28 inches and more thick. Based on an average 54.7-percent recovery from all beds in the county, the recoverable reserves would be 104 million tons. Maps of the Washington, Waynesburg, and Pittsburgh beds are included in the report.

All Brooke County coals are of high-volatile A bituminous rank. Most of them are too high in sulfur, by present standards, to be suitable for manufacturing metallurgical coke.

Ohio County, W. Va.

In this county also the Pittsburgh bed is the one from which comes virtually all of the production.[11/] The reserves in the Waynesburg and Pittsburgh beds aggregated 365 million tons as of January 1, 1954, of which 356 million tons is in areas where the coal is 28 inches and more thick. The average recovery from all beds in the county is 45.4 percent; using this factor, the recoverable reserves are 162 million tons. Maps of the two beds are given in the report.

The Pittsburgh-bed coal is high-volatile A bituminous in rank and cokes strongly. However, because of its high sulfur content in this county, it is unsuitable for making good metallurgical coke under present-day standards.

Overton County, Tenn.

The Wilder bed is the only one in the county from which coal has been produced on a commercial basis.[12/] Estimated reserves in this bed were 19 million tons, as of January 1, 1953, of which 10 million tons is in areas where the bed is 28 inches

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[10/] Dowd, J. J , Provost, J. M., Travis, R. G., Abernethy, R. F., and Reynolds, D. A., Estimate of Known Recoverable Reserves of Coking Coal in Brooke County, W. Va.: Bureau of Mines Rept. of Investigations 5160, 1955, 17 pp.

[11/] Dowd, J. J., Provost, J. M., Abernethy, R. F., and Reynolds, D. A., Estimate of Known Recoverable Reserves of Coking Coal in Ohio County, W. Va.: Bureau of Mines Rept. of Investigations 5171, 1955, 14 pp.

[12/] Williams, Lloyd, Abernethy, R. F., Gandrud, B. W., Reynolds, D. A., and Wolfson, D. E., Estimate of Known Recoverable Reserves and the Preparation and Carbonizing Properties of Coking Coal in Overton County, Tenn.: Bureau of Mines Rept. of Investigations 5131, 1955, 27 pp.

and more thick. Using a determined mining recovery of 58.6 percent, the estimated recoverable reserves are 6 million tons. A map of the bed is included in the report.

Wilder-bed coal is of high-volatile A bituminous rank and is too high in sulfur, under present standards, to be used for making metallurgical coke.

Sequatchie County, Tenn.

The Sewanee bed is the only one in the county from which coal is mined.[13] It was estimated to contain 76 million tons of reserves, as of January 1, 1953, of which 62 million tons is in areas where the bed is 28 inches and more thick. Using a 50.0-percent determined average recovery of coal in mining, the recoverable reserves are 31 million tons. A map of the bed is given in the report.

The Sewanee coal is high-volatile A bituminous in rank and can be used to make metallurgical coke.

Grundy County, Tenn.

Although other beds occur in the county, production comes only from the Sewanee, for which estimated reserves as of January 1, 1953, were 15 million tons. Of this amount, 10 million tons was in areas in which the bed is 28 inches and more thick.[14] The average recovery of coal in mining in the county was found to be 61.3 percent; therefore 6 million tons of coal was estimated to be recoverable. A map of the bed is given in the report.

Sewanee coal in this county is high-volatile A bituminous in rank and makes satisfactory metallurgical coke.

Marion County, Tenn.

Although reserves have been estimated for both the Sewanee and Battle Creek beds in this county, only the reserves in the Sewanee are important.[15] Reserves as of January 1, 1953, were 61 million tons, of which 55 million tons is in areas where the beds are 28 inches and more thick. Using the 60.9-percent recovery determined by the investigation, 34 million tons is considered recoverable. A map of the Sewanee bed is given in the report.

Marion County coal is high-volatile A bituminous in rank, and the Sewanee coal is suitable chemically for making metallurgical-grade coke.

Anderson County, Tenn.

The Pewee, Big Mary, and Windrock beds are the most important in the county. About 73 percent of the known recoverable reserves in the county is in these beds, and 94 percent of the production comes from them.[16] Total reserves in 7 beds, as of January 1, 1953, were estimated to be 170 million tons, including 155 million tons in beds 28 inches and more thick. Using a determined average mining recovery of 57.1 percent, 89 million tons would be recovered in mining. Maps for the Pewee, Big Mary, Windrock, Joyner, Jellico, Coal Creek, and Poplar Creek beds are included in the report.

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[13] Williams, Lloyd, Hershey, R. E., Abernethy, R. F., Gandrud, B. W., and Reynolds, D. A., Estimate of Known Recoverable Reserves and the Preparation and Carbonizing Properties of Coking Coal in Sequatchie County, Tenn.: Bureau of Mines Rept. of Investigations 5136, 1955, 28 pp.

[14] Hershey, R. E., Williams, Lloyd, and Gandrud, B. W., Estimate of Known Recoverable Reserves of Coking Coal in Grundy County, Tenn.: Bureau of Mines Rept. of Investigations 5148, 1955, 16 pp.

[15] Williams, Lloyd, Hershey, R. E., and Gandrud, B. W., Estimate of Known Recoverable Reserves and the Preparation and Carbonizing Properties of Coking Coal in Marion County, Tenn.: Bureau of Mines Rept. of Investigations 5159, 1955, 30 pp.

All Anderson County coals are of high-volatile A bituminous rank. The Pewee, Windrock, Poplar Creek, and some Coal Creek coal meet chemical standards for high-volatile metallurgical coals. The Big Mary bed coal is not suitable because of high ash and high sulfur content.

Coal Investigations in Alaska

The objective of the coal program in Alaska is to insure adequate reserves of minable coal that can: (1) Contribute to the economic growth of the Territory by providing fuel at minimum cost and (2) prevent dependence on waterborne fuel in a national emergency.

The investigation of reserves in the Matanuska coalfield, which supplies the lower rail-belt area, was continued by digging a deep trench across the western end of Wishbone Hill in the vicinity of the Premier mine. This part of the field has been producing a good grade of bituminous coal for many years, but the geology has been obscure at appreciable distance from the old workings. It is expected that further trenching, with confirmation by drilling, will clear up many of the geologic problems. Geologic and engineering data covering exploration work in the Matanuska coalfield are currently being compiled for a forthcoming report of investigations.

In addition to the exploration program, the Anchorage field office keeps in close touch with the Federal Geological Survey and with private exploration work to obtain supplemental information on coal reserves. Private drilling in the Brood Pass area is indicating substantial reserves in moderately thick lignite beds under light glacial cover that may be strippable. In the Beluga field 50 airline miles west of Anchorage, reconnaissance by Bureau engineers confirmed the presence of numerous thick, strippable beds of relatively clean subbituminous coal.

Mining Methods and Practices

Anthracite-Mining Research

Experiments with the Bureau-owned Becorit collapsible steel props were continued, with the objective of developing improved means of roof support for use in mining pillars in thick, flat beds. Much of the remaining reserves in the Northern anthracite field is contained in thick beds, the safe, efficient, and complete recovery of which depends largely upon developing a successful method of roof control.

Conventional roof support is by packing, backfilling, or flushing to fill chamber voids, and the use of wood props, three-piece timber sets, and cogs. Even though this system is practiced most carefully, it has not been successful, because the subsidence, necessary to permit the mine roof to rest upon the packs sufficiently to support it, is too uneven and abrupt to avoid breaking up the roof when wood supports are used.

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[16] Williams, Lloyd, James, Curtis, Gandrud, B. W., and Reynolds, D. A., Estimate of Known Recoverable Reserves and the Preparation and Carbonizing Properties of Coking Coal in Anderson County, Tenn.: Bureau of Mines Rept. of Investigations 5185, 1955, 52 pp.

The ideal roof support is one that will assume a high predetermined and predesigned load with a minimum yield, but, after the design load is reached, it will yield only enough to continue to carry this load without bending. This is accomplished by a yielding action against a calculated resistance. This yielding is very gradual and very slight - only enough to reduce the load upon it below the designed figure. When a group of such supports has been placed in a freshly exposed area, the yielding of one prop (because its load is exceeding the design figure) automatically transmits the overload to other props in the immediate vicinity that have not as yet attained the maximum designed load and are serving as rigid supports. By this action, theoretically all props become loaded to the maximum figure. This action is erratic, the same as with wood timber, but not nearly so pronounced. The result is that the slight movement in the roof that occurs from day to day and week to week is not sufficient to break the roof; instead, a large area of roof rock subsides gradually. The Bureau's two experiments under this project definitely have proved this theory. It has been found that an excellent roof condition prevails, with subsidence approaching 12 inches.

Work with the Bureau-designed, vibrating-blade coal planer described in previous reports was continued to determine whether Pennsylvania anthracite could be mined in this manner and to develop operating data.

The underground testing phase began March 1, 1955, and continued until March 23, when work was suspended because a planer blade broke. While a new blade was being manufactured from a different material, arrangements were completed for purchasing armored-conveyor troughing to replace the wood guide that had been used originally to hold the planer against the coal face and that proved to be inadequate for the purpose.

The tests were resumed October 4 and completed November 25. The theoretical depth of cut was changed from 14 to 9 inches, and the actual depth varied between 6 and 8 inches. The speed of travel averaged 19 feet per minute, with normal chain pull varying between 15 and 20 tons. Momentary peak demands of 40- and 45-ton pull were noted.

Bituminous-Coal-Mining Research

Interest in coal production from thin beds by longwall mining is increasing. Use of the German coal planer in longwall mining was pioneered in this country by the Eastern Gas and Fuel Associates, in cooperation with the Bureau, and the 4 units now in this country have produced over 1 million tons of coal. A fifth planer installation is scheduled for 1956 at a mine in Arkansas where examination showed the feasibility of planer mining.

The first planer installed was moved from the original mine to a nearby operation in West Virginia and produced 221,742 tons in 1955. The second planer produced 88,708 tons during the year at a mine in Pennsylvania where the coal must be blasted to free it from the roof. The hardness of the coal forced suspension of the third planer operation after 5,000 tons was mined. The fourth planer mined 112,792 tons during the year, despite a number of difficulties. Here the coal was harder than was anticipated, and water was present on the face part of the time. A false bottom hampered the effectiveness of roof supports, and it was not possible to keep the planer cutting at the bottom of the bed.

A study of the methods and types of equipment used in the development of mines was begun during the year. The objective of the study was to determine the rate of progress attained with various types of equipment in coal beds of different thickness. The field work was nearing completion at the end of the year. At each mine included in the study, operational details were observed for about 1 week. During this time the operations in a development section and those in a production section were observed in all details, so that results for each type could be compared. Arrangements were made to keep a record of tons produced and man-shifts worked per day in the development section. After a period of 6 to 8 weeks, a return visit was made to measure the advance in development and to get the records of production and man-shifts for the elapsed time. From all data obtained, a report on each operation will be written and submitted to the mine operator for approval and consent to publish.

During the reporting year studies were completed at 12 mines. The varied types of equipment used at the face and in conveying coal from the face in these completed studies are tabulated in table 1. Until all of the studies have been concluded and comparisons made of results, no conclusions can be reached.

Recovery of coal from highwalls of strip mines by means of coal augers is a relatively new coal-mining technique that rapidly has become an important supplement to regular strip-mining methods. The operation consists of drilling large-diameter holes into the coal of strip-mine highwalls. These are drilled at right angles to the face of the highwalls, using large augers that break the coal from the solid and convey it out of the hole and onto a conveyor that loads it into a truck. The general arrangement of the equipment in a strip mine is shown in figure 3; figure 4 shows bits being inserted in a 36-inch cutting head.

Seven auger-mining operations were studied during the year. Operations in 3 States and in 4 different coal beds ranging in thickness from 30 to 96 inches were studied. The auger equipment at these mines was made by three different manufacturers. The diameter of the augers ranged from 26 to 48 inches, and holes up to 208 feet deep were augered. Production ranged from 65 to 503 tons per unit per shift.

Analysis of Coal-Mine-Accident Data

A summary and analysis of reports from coal-producing companies and State mine officials on deaths and injuries to persons in the coal-mining industry for 1951 shows an improvement over 1950, when the best previous record was reported. The improvement was due to a reduction in the injury rate at Pennsylvania anthracite mines from 76.94 per million man-hours of exposure in 1950 to 70.89 in 1951.[17] The frequency rate at bituminous-coal mines (48.72) was virtually unchanged. The total number of injuries, fatal and nonfatal, was 36,338, or 4 percent less than in 1950. Fatal injuries totaled 785, an increase of 142 over 1950. This increase in death toll was due to 5 major disasters (4 at bituminous-coal mines and 1 at an anthracite mine) costing 157 lives. The average severity of all injuries was 188.1 days lost per injury, and the fatality rate for all coal mines was 1.13 per million man-hours of exposure. The largest number of fatalities underground resulted from falls of roof or coal and amounted to 50 percent of the total. Explosions of gas or dust caused 23 percent, and haulage accidents 17 percent, of all underground fatalities.

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[17] Reese, Seth T., Wrenn, Virginia E., and Reid, Elizabeth J., Injury Experience in Coal Mining, 1951: Bureau of Mines Bull. 549, 1955, 73 pp.

TABLE 1. - Equipment used to mine coal and convey it from the face

State	Coal bed	Bed thickness, inches	Mining equipment at face					Haulage for mined coal from face				
			Mobile loading	Continuous mining	Track loading	Cutting	Hand loading	Shuttle cars	Piggy-back	Chain conveyor	Mine cars	Shaking conveyor
W. Va.	Pittsburgh	84	X			X		X				
Do.	do.	84	X			X		X				
Ohio	No. 6	80	X			X		X				
W. Va.	Pittsburgh	73		X				X				
Ky.	Leatherwood	65	X			X			X	X		
Pa.	Upper Kittanning	59			X	X					X	
W. Va.	Sewell	46		X					X	X		
Ohio	No. 6	44	X			X		X				
Pa.	Lower Kittanning	43				X	X					X
Do.	do.	42				X	X					X
W. Va.	No. 3 Elkhorn	36				X	X			X		
Ky.	do.	35	X			X			X	X		

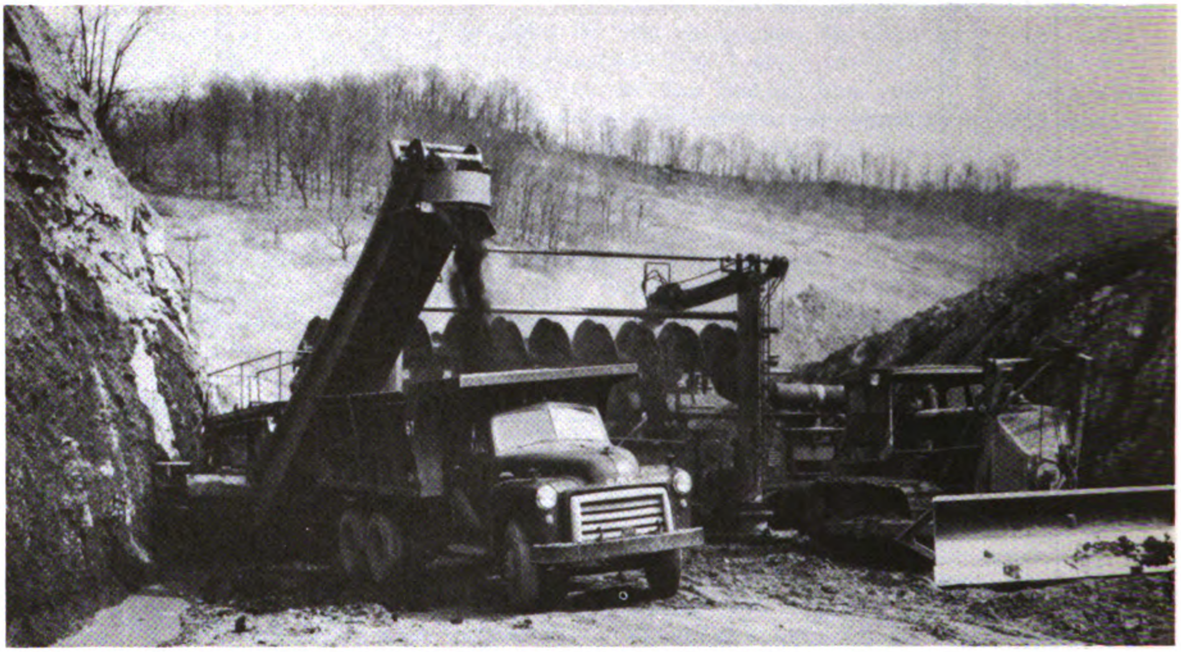

Figure 3. - General arrangement of auger equipment in a strip mine.

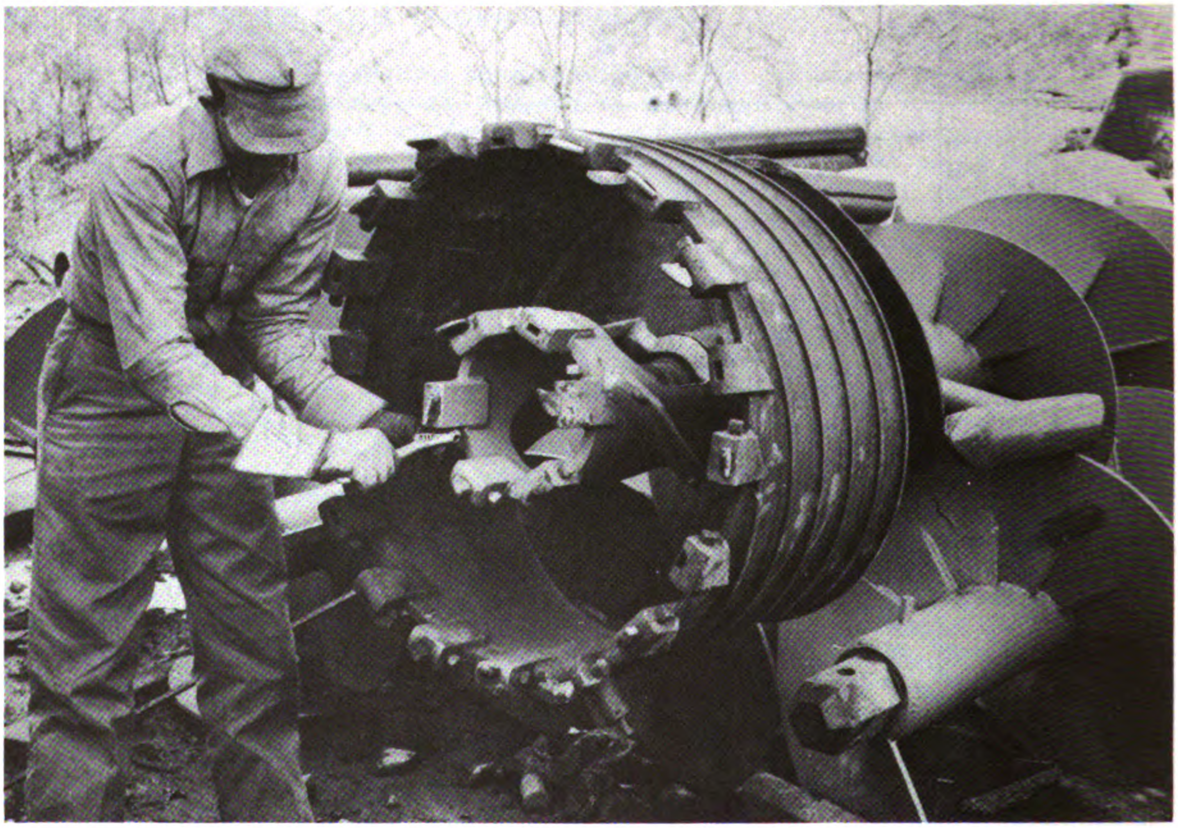

Figure 4. - Inserting bits in 36-inch auger cutting head.

Roof-Fall Fatalities in Coal Mines

During 1954 all of the 174 fatalities from falls of roof, face, or rib in the mining of bituminous coal were investigated, and the circumstances and causes of the accidents were reported.[18/] In 1954, 60 percent of all underground fatalities in bituminous-coal and lignite mines resulted from such falling material, compared with 67 percent for 1953; however, the fatality rate of deaths per million man-hours from this cause was 0.51 in 1954, as compared with 0.49 in 1953 and 0.48 in 1952. Human failure was considered responsible for 93 percent of these fatalities, and only 7 percent were charged to circumstances or risks that apparently could not be foreseen. Seventy-eight percent of the fatalities resulted from accidents within 25 feet of the working face, and 76 percent of the fatalities in this area occurred between the last permanent roof support and the working face. The average length of the comparatively small area from the face to the last permanent support - the most dangerour part of a mine - was 12 feet.

The average dimensions of the pieces of roof rock that fell and resulted in fatalities were: Length, 13 feet; width, 9 feet; and maximum thickness, 1-1/2 feet. The relatively large area of these falls shows that adequate minimum standards for roof control should be adopted and followed in all mines. Coal was loaded mechanically in places where 56 percent of the face fatalities occurred; the remaining 44 percent of the fatalities occurred in places where the coal was hand-loaded.

Twenty-six fatalities occurred in places where roof bolts were used, which represents 15 percent of the total roof-fall fatalities. In only one of the falls did roof-bolt failure cause the accident. In most of these accidents, the fall occurred inby the bolted area; and temporary roof supports, such as safety posts, were not installed. Roof bolts generally offer an improved method of roof control but do not obviate the necessity of providing temporary support inby the bolted area. Indications are that the fatality rate, based on tonnage, from falls of roof in bolted areas is less than that in areas where conventional timbers are used.

Sixty-seven percent of the roof-fall fatalities occurred in solid workings and 33 percent in pillar workings. Reports on 56 underground fatalities in the anthracite mines of Pennsylvania in 1954[19/] revealed that roof falls were responsible for two-thirds of the underground deaths, as in 1953. Haulage accidents caused one-sixth of the fatalities. The remainder were due to strip-mine accidents, moving materials, and other causes.

Recommendations for preventing roof-fall accidents and reducing the number of fatalities from them included: (1) Supervisors should be trained in accident prevention standards; (2) minimum standards for systematic roof support, suited to the conditions in each mine, should be adopted and followed; (3) safe temporary support close to the face should be required before operations are begun; (4) thorough examinations should be made of all working places by competent officials as often as necessary to insure the safety of the workmen; (5) when dangerous conditions are found, they should be corrected promptly; (6) workmen (except those assigned to install supports or inspect the place) should be prohibited from advancing beyond supported roof; (7) permanent supports in active workings should not be removed unless equivalent protection is provided; (8) roof bolting should be adopted when practical;

[18/] Bureau of Mines, Falls of Roof, the No. 1 Killer at Bituminous-Coal Mines, 1954: Bureau of Mines HSS 436, Mineral Industry Surveys, 1955, 9 pp.

[19/] Mather, J. V., Fatalities at Pennsylvania Anthracite Mines, 1954: Bureau of Mines HSS 435, Mineral Industry Surveys, 1955, 18 pp.

(9) roof should be adequately supported to protect men when material is being taken down or roof bolts or crossbars are being installed; (10) employees should attend training classes; and (11) roof supports should not be removed to facilitate loading or to allow movement of equipment unless equivalent protection is provided.

Roof Bolting

Successful bolting depends on being able to make the ground itself an integral part of the support structure, and it cannot be relied on for protection or used alone under other conditions.[20/] The risk of failure of bolted mine roof may now be lessened by using a device known as a roof-bolt compression pad. The Bureau of Mines, in seeking to make roof-bolting installations more reliable, developed a rubber-in-steel bearing plate, and a similar device has been produced by industry that promises to be an effective device warning of impending failure of bolted roof.

Anthracite-Flood Prevention

Reports have been published covering the second and third projects of the five-project comprehensive Conowingo tunnel system.[21/] Each of these proposed projects is a self-contained drainage unit. The second will permit unwatering inundated mines and safeguarding active mines in the Western Middle and Southern fields by pumps discharging into the Schuylkill River, until the main tunnel has been extended to the portal below Conowingo Dam on the Susquehanna River. The third project will perform the same function for the Wyoming Basin of the Northern field and will also handle the water in the Lackawanna Basin, thus permitting the temporary pumping plant provided for this basin in Project No. 1 to be discontinued.

Coal-Mine Ventilation

The effect of changes of atmospheric pressure on methane emissions from extensive worked-out areas of coal mines in several Midwestern States has long caused concern to State and Federal mining departments, as well as to the operating companies involved.

Common mining and ventilation methods employed in mines in these States have resulted in large sealed and unsealed worked-out areas in which considerable quantities of methane are emitted along both intake- and return-air courses.

The common practice is to watch barometric changes very closely and take special precautionary measures during periods of low barometric pressure. Although the hazard is recognized, mining officials have different opinions concerning the effect of barometric-pressure variations on the emission of mine gases. These opinions are based upon scattered personal observations, as no records exist of actual studies to establish such effects.

[20/] Thomas, Edward, Rock Bolting Finds Wide Application: Min. Eng., vol. 6, 1954, pp. 1080-1085.

[21/] Ash, S. H., Dierks, H. A., Kynor, H. D., Lesser, W. H., Miller, P. S., and Romischer, W. M., Flood Prevention in Anthracite Mines, Western Middle and Southern Fields, Anthracite Region of Pennsylvania, Project No. 2: Bureau of Mines Bull. 546, 1955, 37 pp.

Ash, S. H., Dierks, H. A., Kynor, H. D., Lesser, W. H., Miller, P. S., and Romischer, W. M., Flood Prevention in Anthracite Mines, Northern Field, Anthracite Region of Pennsylvania, Project No. 3 (Wyoming): Bureau of Mines Bull. 547, 1955, 35 pp.

An initial study during February and March 1954 in the Orient No. 2 mine of the Chicago, Wilmington & Franklin Coal Co., West Frankfort, Ill., to determine the effect of atmospheric pressure changes on gas emissions from extensive worked-out areas indicated the following, although the data obtained were not as complete as desired.22/ (1) The four underground areas selected for study proved to be comparatively innocuous, owing to breathing action caused by barometric changes; (2) breathing action from sealed areas greatly increased the methane content of main return-air currents during barometric lows; (3) small changes in barometric pressure do not effect material changes in the gas content of mine-air currents; and (4) the rate at which a seal "bleeds" air or air-gas mixtures is determined by its construction and the pressure differences on it.

Determining Explosibility of Mine-Fire Atmospheres

Mine fires are usually extinguished by constructing seals in the approaches to the areas to exclude air from the fire areas. Information obtained on the composition of atmospheres behind fire seals is helpful in determining if a fire has been extinguished or if there is danger of an explosion when an area is being sealed or unsealed. The progress of a mine fire can best be determined by studying the percentages of carbon dioxide, carbon monoxide, and hydrogen in the atmosphere behind the seals. A sealed fire area should not be reopened until carbon monoxide has disappeared and carbon dioxide is static. Analyses of a mine-fire atmosphere will show whether it is explosive, if it is capable of becoming explosive when mixed with air, or if it can be made nonexplosive by mixture with an inert gas. These factors can be computed mathematically or determined graphically. The graphical solution can be made in minutes and is extremely helpful while a fire is being fought.23/

Detecting and Determining Carbon Monoxide

In industrial and domestic daily life carbon monoxide is the toxic gas encountered most frequently. Hazardous concentrations of carbon monoxide may be present in the vicinity of many industrial operations, such as metallurgical processes, coke ovens, and fuel- and synthesis-gas-producers and may be encountered in confined spaces, such as dwellings, garages, workshops, vehicular tunnels, and mine workings, in connection with the use of heaters, boilers, internal-combustion engines, after blasting, and after fires and explosions. Evaluation and control of the hazards caused by the gas depend primarily on the analytical detection of its presence and determination of its concentration. The instruments and procedures commonly used for determining carbon monoxide are of interest to many persons of varying degrees of technical training.24/

The National Bureau of Standards calorimetric carbon monoxide indicator is a sensitive, hand-operated device for rapid detection and determination of low concentrations of carbon monoxide in air. It is sensitive to 0.001 percent carbon monoxide and will detect 0.01 to 0.04 percent at ground level and 0.0025 to 0.05 percent aboard aircraft in 1 to 5 minutes. The indicator is operated by aspirating an air sample through a small glass tube containing silica gel impregnated with

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22/ Dornenburg, D. D., O'Connor, J. A., and Harris, E. J., Effect of Changes of Atmospheric Pressure on Gas Emissions From Worked-Out Areas in an Illinois Coal Mine: Bureau of Mines Rept. of Investigations 5147, 1955, 21 pp.

23/ Shaw, J. F., A Graphic Method of Determining the Explosibility Characteristics of Mine-Fire Atmospheres: Bureau of Mines Bull. 551, 1955, 11 pp.

24/ Beatty, R. L., Methods for Detecting and Determining Carbon Monoxide: Bureau of Mines Bull. 557, 1955, 34 pp.

palladous sulfate and ammonium molybdate. The color change of the gel from light yellow to progressively darker green is matched with a series of color standards to indicate the carbon monoxide in the sampled air.

The hoolamite or activated iodine pentoxide carbon monoxide detector may be used to determine semiquantitatively 0.1 to 1.0 percent of carbon monoxide in air contaminated with smoke and fumes from fires and explosions. The carbon monoxide recorder is an instrument for continuously sampling and recording low concentrations of carbon monoxide in air; it is used extensively to record and control exposure to carbon monoxide in vehicular tunnels. The carbon monoxide indicator is a portable instrument used for continuous determination of carbon monoxide in air under various circumstances of suspected contamination. The portable indicator operates on the same principle as the carbon monoxide recorder. Carbon monoxide is oxidized in a hopcalite cell, and the heat of oxidation generates an electric potential in a thermocouple system connected to a millivoltmeter graduated in terms of carbon monoxide concentration from 0.0 to 0.15 percent. The carbon monoxide alarm may be used to sample and analyze continuously air that may be contaminated by carbon monoxide. It operates on the same principle as the recorder and the portable indicator.

The pyrotannic-acid colorimetric method is used to determine the carboxyhemoglobin saturation in samples of blood and the concentration of carbon monoxide in air by using blood-reagent solutions that have been equilibrated with air samples. The iodine pentoxide method is the standard laboratory procedure for sensitive, accurate, chemical determination of carbon monoxide in air and in various gas mixtures. The method is based upon measurements of the iodine liberated by the oxidation of carbon monoxide by heated iodine pentoxide. The analytical range is from 0.0005 to about 1.0 percent. Low concentrations of carbon monoxide may be determined by the palladous chloride-phosphomolybdic acid-acetone colorimetric method. The color intensity of molybdenum blue formed by oxidizing action on carbon monoxide is measured with a photoelectric colorimeter. Infrared gas analyzers may be used to determine and record continuously concentrations of carbon monoxide. The specific absorption of infrared radiation by carbon monoxide and other molecular gases is a distinctive physical property. Carbon monoxide may be determined by gas-volumetric methods of analysis.

Testing Electrical Equipment

To enable the mining industry to be kept informed as to the availability of equipment developed by mining-equipment manufacturers in their constant endeavor to keep a step ahead of changes in mining techniques, the Bureau of Mines periodically issues lists of equipment approved since the last previous listing.

The present list[25] covers those approvals issued during calendar years 1953 and 1954. It supplements Bulletin 543, Permissible Mine Equipment Approved to January 1, 1953. The procedure, established in Bulletin 543, of numbering items in each category consecutively to indicate the chronological order of the approvals has been continued. Testing and approval are conducted under the provisions of the schedules administered by the Electrical-Mechanical Branch. These schedules are revised and amended to keep them current with the improvements and new developments in processes and equipment. The latest revisions of the schedules involved are: Schedule 2F (Procedure for Testing Junction Boxes and Electric Motor-Driven

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[25] Kearns, R. A., and Brunot, H. B., Permissible Mine Equipment Approved During the Calendar Years 1953-54: Bureau of Mines Inf. Circ. 7722, 1955, 12 pp.

Equipment for Permissibility); Schedule 6D - Amended (Permissible Electric Cap Lamps: Procedure in Testing, Fees Charged, and Requirements for Approval); Schedule 7C - Amended (Flame Safety Lamps: Requirements for Permissibility, Tests Made,and Fees Charges); Schedule 8C - Amended (Portable Methane Detectors, Requirements for Permissibility, Tests Made, and Fees Charged); Schedule 9B - Amended (Permissible Telephones and Signaling Devices: Requirements for Permissibility, Tests Made, and Fees Charges); Schedule 10C - Amended (Electric Mine Lamps, Other Than Cap Lamps; Requirements for Permissibility, Tests Made, and Fees Charges); Schedule 12D - Amended (Single-Shot Blasting Units); Schedule 16B - Amended (Procedure for Testing Multiple-Shot Blasting Units); Schedule 22 - Amended (Procedure for Testing Diesel Mine Locomotives for Permissibility); Schedule 25A (Procedure for Testing Dust Collectors for Permissibility for Use in Connection With Rock Drilling in Coal Mines).

Electrical apparatus used in mines is studied to determine whether its operation and use might introduce a hazard. From this study, certain safety standards have been developed. These standards, established with the aid of both the manufacturer and user of such apparatus, are published in schedules, and apparatus that complies with the requirements set forth in these schedules is classified as "permissible." This term is defined to designate apparatus as "completely assembled and conforming in every respect with the design formally approved by the Bureau of Mines." In the judgment of the Bureau, the manufacturers of such apparatus have adhered to certain minimum standards of safety in supplying a product that, when properly used and maintained, is safe for operation in mines where flammable gas or coal dust may accumulate.26/

Permissible mining equipment was introduced in the United States in 1914 but was not generally used for many years. During the period in which the Federal Mine Safety Code was used as the basis for Federal coal-mine inspections, design and production of permissible equipment have been greatly stimulated. Over 60 manufacturers have been granted formal approval of their products since the first permissible machine was placed on the market. Thus, some manufacturers have accumulated over 30 years' experience in designing equipment to meet the requirements of the Bureau as contained in the published schedules.

Preventing Mine Fires and Explosions

Investigation reports are available on 51 mine fires of electrical origin between August 15, 1949, and August 16, 1954. Nine other fires in the same period were suspected of being of electrical origin. Every mine fire is a potential hazard to underground workmen; it is remarkable that only six lives were lost as a result of these fires, since the injury list was very high.27/

Modern mining methods, with increased power demands and extended distribution systems underground, involve greater electrical hazards and possibilities of costly production delays as a result of inadequate electrical service. Improvements are desirable in electric-power-distribution facilities in many underground coal mines.

Fire hazards from underground power-distribution systems are at a minimum only where the system is designed in accordance with accepted good engineering practices and so protected that a direct circuit at any place on the system will cause power

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26/ Gleim, E. J., James, R. S., and Brunot, H. B., Explosion-Proof Design and Wiring for Permissible Mining Equipment: Bureau of Mines Bull. 541, 1955, 40 pp.

27/ Brown, C. L., Preventing Coal-Mine Fires Caused by Electrical Equipment: Bureau of Mines Inf. Circ. 7716, 1955, 20 pp.

to be removed automatically from the affected part of the system. The cost of minimizing fire hazards where they exist on underground power systems will vary for different mines, depending upon the magnitude of the deficiency. The manner in which the correction is made is not always limited to the application of automatic protective devices. The most practical solution, in any event, requires proper engineering knowledge of the problem. Common methods of providing adequate short-circuit protection where such deficiencies exist are: (1) Increase current-carrying capacity of power circuits by adding copper or improving track bonding as required; (2) maintain substations as close to load centers as is practicable; (3) isolate substations where feasible; (4) reduce settings of automatic overcurrent devices to conform to safe current-carrying capacity of the circuit; (5) check the locations of protective devices for maximum effectiveness, mine development often requires relocation of devices; (6) where immediate improvements cannot be made, reduce the length of power circuits by discontinuing work in remote areas; and (7) install automatic overcurrent devices as required.

Several recent ignitions of firedamp (a mixture of methane and air) in American coal mines have been attributed to frictional heating when hard rocks were struck during cutting and drilling. Although none of these ignitions caused severe explosions, they call attention to a problem that can become increasingly significant as mechanized mining is developed further.28/ The problem of frictional heating is not new in coal mining. In Europe, particularly in England, it has been a concern for more than a century. During the last 15 to 20 years, frictional ignitions have been a major cause of explosions in British mines. The greater number of such ignitions in England may be due to the special character of the coal-measure strata, the mining methods, the rate of gas emission, and possibly its composition, and less effective ventilation near the face. In general, flammable mixtures of gas are ignited by frictional heating in two ways: (1) A small volume of gas is heated to the ignition temperature by direct contact with a hot part of one of the rubbing surfaces; or (2) small heated particles, commonly termed frictional sparks, are projected into the gas mixture. These frictional sparks are small, hot particles of solid matter torn from one of the larger pieces. The particles may be inert, in which case their temperature is limited by their melting point; or they may be chemically active, and their temperature is augmented by oxidation. The ability of sparks to ignite firedamp is greatly restricted by the short period of contact with the gas, owing to cooling by convection, and to the "ignition-lag" period of the gas that varies with composition and with temperature. All of these factors make flying sparks generally less dangerous than sparks whose paths are obstructed by a thermal insulator at an early stage. The surfaces whose friction or impact has resulted in dangerous heating or sparking include rocks against rocks, metals (especially steel) against rocks, steel against steel, and light metals against steel. The mine rock most likely to create a hazard is sandstone, while iron pyrite ranks next. A few ignitions have been reported when hard shales and ironstone (a stratified iron ore, generally siderite) were struck by steel.

To avoid ignitions, gas accumulation and frictional heat or sparks must be prevented. Measures proposed for this purpose include many based on European experience and research, some of which have no practical application to mechanized mining conditions.

The most important recommendations probably relate to the provision of adequate ventilation at the coal face, the use of enough air and water in cuts, frequent checking for gas in the atmosphere, the design and use of mining machines to

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28/ Hartmann, Irving, Frictional Ignition of Gas by Mining Machines: Bureau of Mines Inf. Circ. 7727, 1955, 17 pp.

facilitate rapid removal of gas, avoidance of overheating of cutting and other machines, and avoidance, insofar as possible, of hard rocks during cutting and drilling.

All pipelines used to transport liquid fuels over coal mines are potentially hazardous because of the explosion and fire hazards that arise from seepage of gasoline into underground mines. Once gasoline has entered mine workings, it is extremely difficult to remove, especially when pools may exist in abandoned areas.29/ In two small Pennsylvania mines in February 1953, gasoline from a pipeline leaked into the workings, where the vapors ignited and persons were burned. According to officials of the pipeline company, corrosion and subsequent leaking of their pipelines arise from four main causes: (1) Natural oxidation; (2) electrolysis; (3) chemical reaction of soils and water; and (4) chemical reaction caused by certain bacteria.

Very small concentrations of gasoline can be detected by the sense of smell. Experiments carried out by the Bureau of Mines have shown that the odor of gasoline is noticeable in inhaled air at a concentration of 0.03 percent. The investigators found that they could detect gasoline vapor by smell long before any indication was registered on a testing instrument.

Safety Training and Education

First-aid training has been an important activity of the Bureau of Mines since its establishment. From 1910 to 1926 first aid was taught only by trained Bureau of Mines employees; consequently, only a comparatively few persons could be trained. In 1926, however, cooperative first-aid training was initiated; this provided the means to train the entire personnel of mines or plants within a short time in a single campaign. Many mines and plants have taken advantage of the cooperative plan, whereby keymen in a mine or plant are given intensive training in first aid by a Bureau of Mines instructur, in addition to the initial training received. As instructors, under the supervision of a Bureau of Mines instructor, these men train classes of persons employed in the mineral industries in the standard Bureau of Mines course in first aid. A mine, plant, or company in the mineral industries that completes training its entire personnel in the Bureau of Mines standard first-aid course receives a large, engraved certificate showing that 100 percent of the employees have been so trained. Local labor organizations having all the members trained also receive the 100-percent certificate.30/

Since its organization in 1910, the Bureau of Mines has assisted mining companies and other organizations connected with the mineral industries in conducting first-aid and mine rescue contests by providing instructors to train teams, judges, and other officials; by furnishing problems and outlines for working them, score cards, and contest rules; and by assisting in any way they were requested.31/

In the 1953 National First-Aid and Mine Rescue Contest 58 teams from 9 States participated - 14 teams from 6 States in mine rescue and 47 teams from 9 States in

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29/ Walker, W. D., Jr., and Dumire, J. H., Coal-Mine Hazards From Overlying Gasoline Pipelines: Bureau of Mines Inf. Circ. 7708, 1954, 14 pp.

30/ Moschetti, A. C., and Fritts, G. B., 100-Percent Cooperative First-Aid Training, Mines and Quarries, Colorado Fuel & Iron Corp., Colorado and Wyoming: Bureau of Mines Inf. Circ. 7712, 1955, 10 pp.

31/ Warncke, R. G., National First-Aid and Mine Rescue Contest, Fort Wayne, Ind., Sept. 29 and 30, and Oct. 1, 1953: Bureau of Mines Inf. Circ. 7710, 1955, 82 pp.

first aid. The contest was held under the auspices of the Federal Bureau of Mines and the Joseph A. Holmes Safety Association and was sponsored by the National Coal Association, United Mine Workers of America, and various State mining departments.

The nationwide nature of the Bureau's work gives its representatives a far better opportunity to become acquainted with matters pertaining to health and safety in the mines of the United States than members of any other agency; hence, it is appropriate that the Bureau of Mines should assemble and issue to the mining public its findings as to health and safety in the industry after its experience of many years. A safety course for bituminous-coal-mine officials and persons who aspire to become officials has been formulated, and its component parts have been published so that they may be utilized, not only by Bureau of Mines instructors in giving the course, but also by officials of mining companies and others who take the course and wish to keep well-informed then and afterward. Recent revisions of the sections of the text have presented advances in technology and practice in hoisting and haulage,[32] prevention and control of mine fires and explosions,[33] safe methods and practices in handling and using explosives,[34] protection against miscellaneous hazards such as machinery, falls of persons, flooding, and information on sanitation and health.[35]

Foreign Coal Investigations and Technical Assistance

As part of the Government's program of technical assistance to underdeveloped areas, coal projects were continued in Afghanistan, Colombia, Nepal, India, Pakistan, and the Philippines under sponsorship by the International Cooperation Administration (ICA). Most of this work is intended to increase local coal supplies, because the necessity of importing fuel is a heavy burden on the economy of countries that are straining to increase their industrial capacity. The training of foreign nationals, both in their home countries and in the United States, is an important phase of the program.

Afghanistan

The two Bureau engineers in Afghanistan have been training and instructing personnel to operate the country's two active coal mines, Karkar and Ishpushta. The mining conditions at both are extremely difficult, because of steeply dipping, irregular beds. Heretofore the chief source of power at the mines has been diesel engines, but a powerline to the Karkar mine is now being constructed. The new supply of power at Karkar will enable the production there to be raised, but small reserves at Ishpushta limit expanded production.

An important part of the program during 1955 was planning and exploration for development of the Dara-Suf area, which is believed to contain substantial reserves. This isolated region, northwest of Kabul, is at present accessible only by a roundabout route from the north. A new southern route through the difficult mountainous terrain has been selected. A small amount of equipment for this access road has been ordered, but the major part of the road building will be done by local Afghan hand labor.

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[32] Accidents from Hoisting and Haulage in Bituminous-Coal Mines: Bureau of Mines Miners' Circ. 49, (1954 revision) 1955, 88 pp.

[33] Bureau of Mines, Explosions and Fires in Bituminous-Coal Mines: Miners' Circ. 58 (1954 rev.), 1955, 86 pp.

[34] Bureau of Mines, Explosives Accidents in Bituminous-Coal Mines: Miners' Circ. 58 (1954 rev.), 1955, 86 pp.

[35] Bureau of Mines Miscellaneous Accidents in Bituminous-Coal Mines: Miners' Circ. 60 (1954 rev.), 1955, 85 pp.

At the two operating mines a safety course imparting the principles of fire prevention, mine rescue, and first aid has been conducted throughout the year. A total of 21 mine managers, foremen, and operators has attended regularly. A text on coal-mine safety is now being prepared as a future reference work. The engineer conducting the course and preparing the text is emphasizing standard Federal Bureau of Mines techniques.

In addition to production problems, coal utilization in Afghanistan presents a number of difficulties. The chief of these is the excessive amount of fines produced in mining, with the further degradation that takes place in handling before the coal reaches the Kabul market. A study is now underway of briquetting processes and equipment to replace the present hand presses, in which the briquets are formed with the use of a small amount of clay and starch as binder.

Colombia

As part of a general development plan for the Cauca Valley, a Colombian Government corporation is erecting a coal-preparation plant in the vicinity of Cali to produce 125 tons of prepared coal per hour. Three of the mines in the valley are being reequipped to increase their production enough to supply the new washing plant. The largest part of this equipment, including safety apparatus, ventilating fans, hoists, and locomotives, is being ordered from France. A Bureau of Mines engineer was occupied continuously during the year in assuring proper use of this material as it is received and in giving advice on efficient and safe mining methods.

India

Assistance on development of the open-cast mine at Bermo, operated by the Indian Government, was concluded during the year. In his final report the Bureau engineer recommended that careful consideration be given to diesel-truck haulage, instead of the favored electric-track-haulage system.

At South Arcot, Madras State, in South India, a Bureau of Mines coal-mining engineer was stationed during the first half of the year as advisor on open-pit development of the lignite deposit. The South Arcot lignite is the chief source of fuel available in South India and is one of India's major coal deposits. A production of 1 to 3 million tons of lignite per year is envisaged. This would require stripping 4 to 12 million cubic yards of overburden. Various mining schemes are under study, including most opencut-mining and ore-handling methods. Indian engineers favor rail haulage out of the pit, although the use of trucks is also under study, in spite of the necessity of importing liquid fuel. The use of Sauerman tower excavators has been studied and tentatively rejected because of the great depth of the material to be mined.

Concurrent with study of mining methods at South Arcot, attention is being given to various utilization proposals. Six alternative proposals were advanced by British consultants to the Indian Government for utilizing 1 to 6 million tons of lignite a year, employing German technology. During 1955 the Bureau of Mines, both in India and in the United States, gave considerable attention to these proposals and to alternative proposals using American techniques. One essential difference between the latter and the German system is that in the proposed American scheme all the lignite is dried in a central drying plant, whereas the German technology uses individual drying units for each operation of combustion, briquetting, and carbonization. At the end of 1955 approval was received for two Bureau engineers to proceed to South Arcot to study local conditions and to recommend the first stage of pilot-plant construction. It is expected that a process for drying in the entrained and fluidized state, similar to that employed at Rockdale, Tex., will be advocated. A discussion of the preparation and utilization properties of the South Arcot lignite is given later in this report.

Nepal

The Bureau program in this country is in its initial stage and is concerned chiefly with investigation of reported deposits of minerals. A deposit southeast of Katmandu, near Barahachhetra, was examined; it turned out to be carbonaceous shale and definitely had no value as fuel. Other coal deposits are reported in Nepal, and some of them were examined during 1955 by a coal-mining engineer on temporary assignment for ICA. His report was unfavorable. The Bureau representative has planned future examinations of similar deposits during the coming year.

Pakistan

The Bureau has been giving advice regarding construction of a fertilizer plant at Daud Khel that is to produce 50,000 tons of ammonium sulfate yearly. Operation of this plant will require approximately 350 tons of coal a day that is expected to come from the Makarwal colliery. This coal output will be largely additional to the approximately 150 tons a day now being produced, although some cutback in other uses is planned. The Bureau has had no direct connection with the Makarwal development but has continued to study and urge upon ICA and the Pakistan Government the vital importance of having this mine operating at the required rate when the fertilizer plant is ready.

Philippines

The objectives of Bureau work in the Philippines are: To investigate the coal-bearing areas by diamond drilling, in order to establish minable reserves in preparation for possible future mining; to undertake thorough laboratory testing of coals from different sources in the Philippines; and to investigate the utilization and marketing of Philippine coals. The work is done in cooperation with the Philippine Bureau of Mines; American support will be withdrawn as soon as the Philippine Government is able to continue alone.

During 1955 the two Bureau engineers concentrated their assistance on practical advice on drilling to the Philippine Bureau of Mines field team at the Lumbog mine area of Mindanao. Two drills were being operated in that vicinity, and three additional holes were planned. Completion of this work will allow an accurate estimate of reserves of the highest rank coal in the Philippines. The coal reserves adjacent to the Lumbog mine were tentatively estimated in December 1955 as about 500,000 tons, enough for about 10 years of production at the current rate. On Bataan Island, off the southeastern coast of Luzon, drilling was in progress throughout the year with the continuous assistance and advice of the Federal Bureau of Mines. Unfortunately, results of the drilling were disappointing, and the work has been abandoned.

A forthcoming Bureau report will present the results of the original field investigations in the Philippines; the tests of carbonizing properties of higher rank Philippine coals performed at Pittsburgh, Pa.; and the work on utilization of Philippine noncoking coals at the Denver Station of the Bureau of Mines.

Aid to the Philippine Bureau of Mines on determination of coal reserves was terminated at the end of 1955. Future assistance in this field will depend on an evaluation of the role of solid fuels in the Philippine economy. At present, only informal assistance is being given by other Bureau personnel assigned to minerals work in the Philippines.

COAL PREPARATION AND STORAGE

Improved coal-preparation methods can make a major contribution toward conservation of coal resources by permitting marginal coals to be upgraded to meet metallurgical standards and by allowing the mining of impure beds that otherwise would be wasted. Thus, much of the Bureau's research in this field is intended to develop better means of tailoring coals to meet market requirements at minimum cost and with minimum waste. Another major phase of the work is the conduct of washability studies and cleaning trials to determine the extent to which various coals can be improved by cleaning.

Coal Washing

Arizona

To determine the washability characteristics of the substantial reserves of subbituminous and bituminous coals on the Hopi and Navajo Indian Reservations in the Black Mesa field, Ariz.,[36] samples of coal from six mines in Navajo and Coconino Counties were float-and-sink tested. The ash content of 2 of the raw-coal samples analyzed about 5 percent, and further reduction in ash content by mechanical cleaning would be difficult and uneconomical. The remaining four coals were amenable to substantial improvement by mechanical cleaning. Many deposits appear to contain extensive reserves, much of which could be recovered by strip-mining methods. It is most unlikely that these deposits will be developed significantly within the foreseeable future, owing to the isolated area in which the coal is found and lack of market.

Montana

Detailed information on the cleaning of Montana coals has been made available to assist the industry in meeting present and future market requirements without sacrificing the public interest in conservation.[37] Coals from all principal-producing fields in Montana were covered in the investigation, which included washability examinations, pilot-plant cleaning trials, and an analysis of washery operation.

The coals examined from the Red Lodge, Bull Mountain, and Forsyth fields are comparatively easy to clean. Ash contents of 5 to 10 percent can be obtained by eliminating the nominal percentage of impurity contaminating them. The coal from the Great Falls field, however, cannot be cleaned readily to less than 14 percent ash, even though a high percentage of impurity is rejected.

Pennsylvania Anthracite

The modern, high-capacity, Baum-type jig used so widely in cleaning bituminous coal has not been adopted in the Pennsylvania anthracite region, where small-capacity, piston jigs are used to treat closely sized coal. The substantial reduction in cleaning costs that could be achieved by treating a wide range of sizes in the Baum-type jig prompted the Bureau to investigate its effectiveness in cleaning anthracite. The results of this investigation have been prepared for publication as a Report of Investigations.

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[36] Crentz, W. L., Washability Characteristics of Coals From the Black Mesa Field, Arizona: Bureau of Mines Rept. of Investigations 5116, 1955, 20 pp.

[37] Geer, M. R., and Yancey, H. F., Cleaning Characteristics and Cleaning Tests of Montana Coals: Bureau of Mines Rept. of Investigations 5103, 1955, 47 pp.

In addition to these cleaning trials, the study of anthracite crushing practices has continued. Various crushing methods are being compared to determine which provide the highest recovery of the finer sizes of anthracite now demanded by the market. Study to date has covered nonroll crushers and has included impact, jaw, ring, and gyratory types. Initial work was concerned with obtaining maximum recovery of Buckwheat and Rice (9/16- by 3/16-inch) sizes when the larger, hardest anthracites were crushed. This year has seen the work expanded to determine the maximum obtainable recovery of all the finer sizes when coals that typify the extremes of hardness (friability) encountered in the region are crushed. Overall results of comparable tests have shown that the gyratory- and ring-type crushers will yield the maximum recovery of Buckwheat and Rice sizes from crushing hard anthracites. The gyratory-type crusher produced 13 percent less Buckwheat and Rice sizes when the friable coals are reduced, compared with the 62-percent recovery obtained with the hardest anthracites. However, either the hard or friable coals, by proper choice of machine settings, can be crushed in the gyratory-type crusher to produce 75 to 85 percent of Buckwheat No. 1 through Buckwheat No. 5 (3/64-inch by 30-mesh) sizes. The performance of the ring-type crusher when friable anthracite is reduced has yet to be determined. It is expected that study of roll-type crushers to determine their maximum application in anthracite-crushing practices will begin after the work with non-roll-type machines is completed.

Alaska

A comprehensive performance test was made in a new cleaning plant built in Alaska to recover coal from an unusually dirty bed formerly wasted in a stripping operation. The data obtained will assist materially in evaluating the technical and economic feasibility of treating similar coals encountered elsewhere, or in the more widely applicable problem of recovering coal from refuse.

This plant employs the dense-medium process, which generally provides recovery efficiences approaching 100 percent when applied to coals of ordinary character. With this unusually difficult coal, however, the recovery efficiency obtained was substantially lower, indicating the necessity for considering this factor in planning similar operations.

Coal Recovery From Refuse

An investigation of the feasibility of recovering salable coal from washery waste has been completed.[38] Interest in this subject stems from the fact that many older preparation plants in the country are operating well below their original efficiency, with a resultant loss of coal that constitutes an economic burden on the operators and a resource waste to the public. The loss of coal occurs primarily because these plants must cope with treatment problems that are much more difficult than those for which they were designed. The constant demand for greater underground productivity has dictated full-seam mining, with an attendant increase in the proportion of dirt contaminating the raw coal. Also, in many instances the depletion of cleaner beds has forced transfer of mining operations to beds containing more impurity. At one mine studied the loss of salable coal has increased sixfold during the life of the washery because of these factors.

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[38] Geer, M. R., Fennessy, F. D., and Yancey, H. F., Recovery of Coal From Washery Refuse; Some Examples of the Effect of Continuous Mining Machines on Preparation Problems: Bureau of Mines Rept. of Investigations 5152, 1955, 45 pp.

In the course of the investigation, samples of refuse from three jig washeries were used for cleaning trails in a dense-medium pilot plant to demonstrate application of this process in recovering coal from waste. These tests demonstrated that coal could be separated from impurity with the same degree of sharpness obtained in treating raw coal by dense medium. However, the recovery efficiency achieved in treating refuse averaged distinctly lower than was possible with raw coal because of the greater proportion of intermediate-density material and heavy impurity to be dealt with.

Industrially significant byproducts of this and earlier work with the dense-medium pilot plant were development of an improved device for continuously measuring specific gravity of dense mediums[39/] and an evaluation of various methods of appraising coal-washing results.[40/]

Additional information on the recovery of coal from waste was made available in a report[41/] describing operation of a commercial plant treating 175 tons per hour of refuse derived from the Pittsburgh bed. An exterior view of this plant is shown in figure 5 and the flowsheet in figure 6. The plant treats 4- to 3/8-inch material in a dense-medium vessel, the medium for which is obtained by pulverizing rock handpicked from the plant feed.

Samples of the feed to the separating bath, the washed coal, and the final refuse were subjected to specific-gravity analyses to determine the effectiveness of the separation between coal and impurity. When the plant was operating on weathered material that had been stored 2 years, a washed coal, analyzing 8.5 percent ash, was obtained from a feed analyzing 57 percent. The yield of washed coal was 27.5 percent, and the recovery efficiency was 86.8 percent. Treating fresh refuse at the same specific gravity, a washed coal of 8.1 percent ash was obtained at a yield of 20.8 percent and a recovery efficiency of 89.3 percent. The fresh refuse analyzed 67 percent ash and thus yielded less clean coal.

Kerosine Flotation

It is generally agreed that the most difficult part of coal preparation is cleaning and dewatering the very fine sizes. Since this material cannot be treated satisfactorily by processes that depend on differences in the specific gravities of the coal and associated impurities, some other property (either chemical or physical) must be utilized, and present indications are that processes depending on differences in wetting characteristics are most suitable for commercial exploitation. Coal-preparation research at the Southern Experiment Station is therefore concerned principally with a study of the theory and practice of coal flotation. Results of this study should lead to more efficient preparation methods, which, in turn, should contribute materially to the preservation of our coal reserves.

[39/] Geer, M. R., and Klein, M. J., Device for Continuously Measuring Density of Unstable Suspensions: U. S. Patent 2,708,360, 1955, 3 pp.

[40/] Yancey, H. F., and Geer, M. R., Some Practical Aspects of Appraising Coal-Washing Results: Bureau of Mines Rept. of Investigations 5153, 1955, 15 pp.

[41/] Palowitch, E. R., Jolley, T. R., and Sokaski, M., The Use of Pulverized Rock as Dense Medium for Salvaging Coal From Pittsburgh-Bed Refuse: Bureau of Mines Rept. of Investigations 5184, 1955, 33 pp.

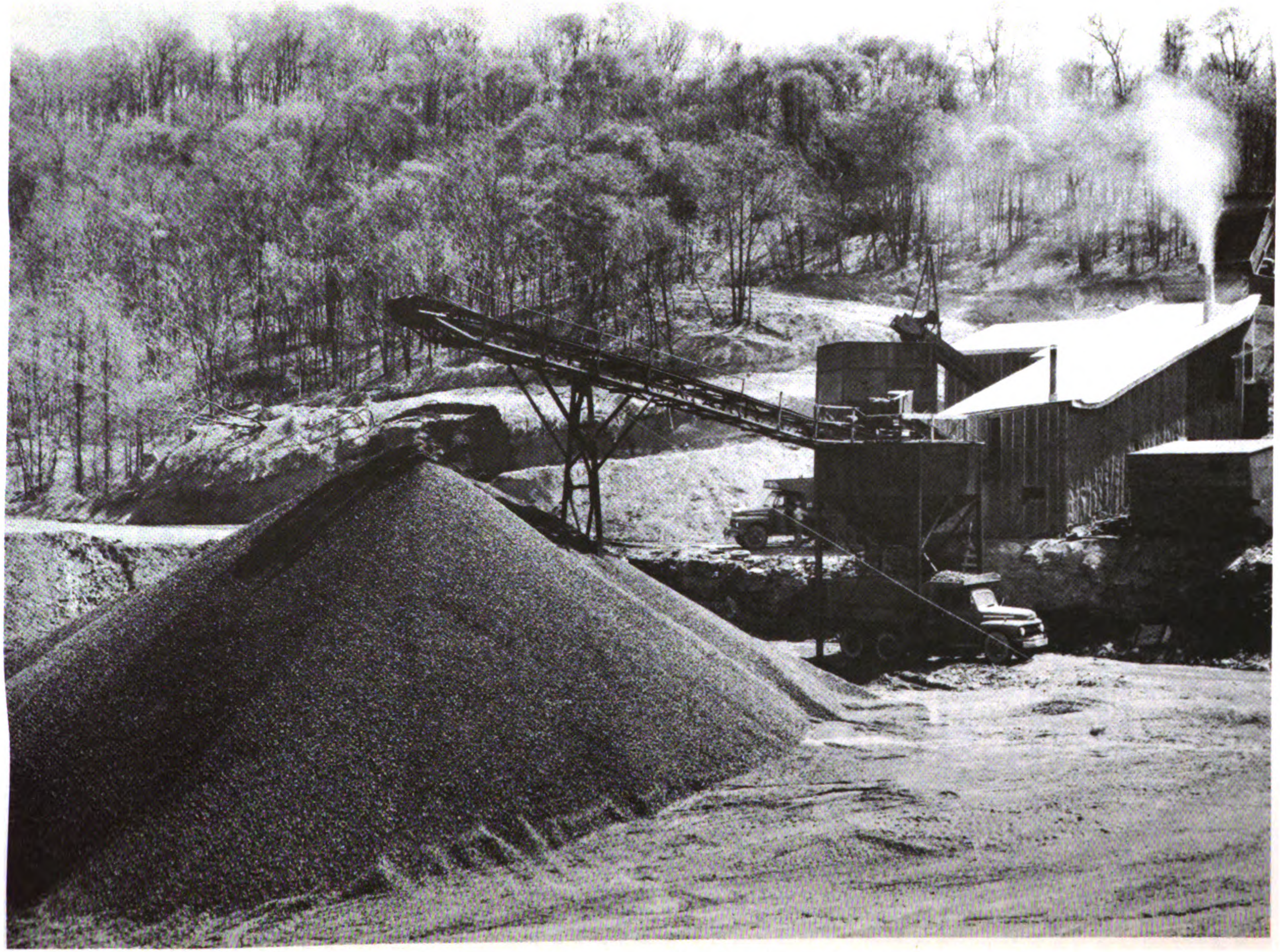

Figure 5. - Plant salvaging coal from Pittsburgh-bed refuse.

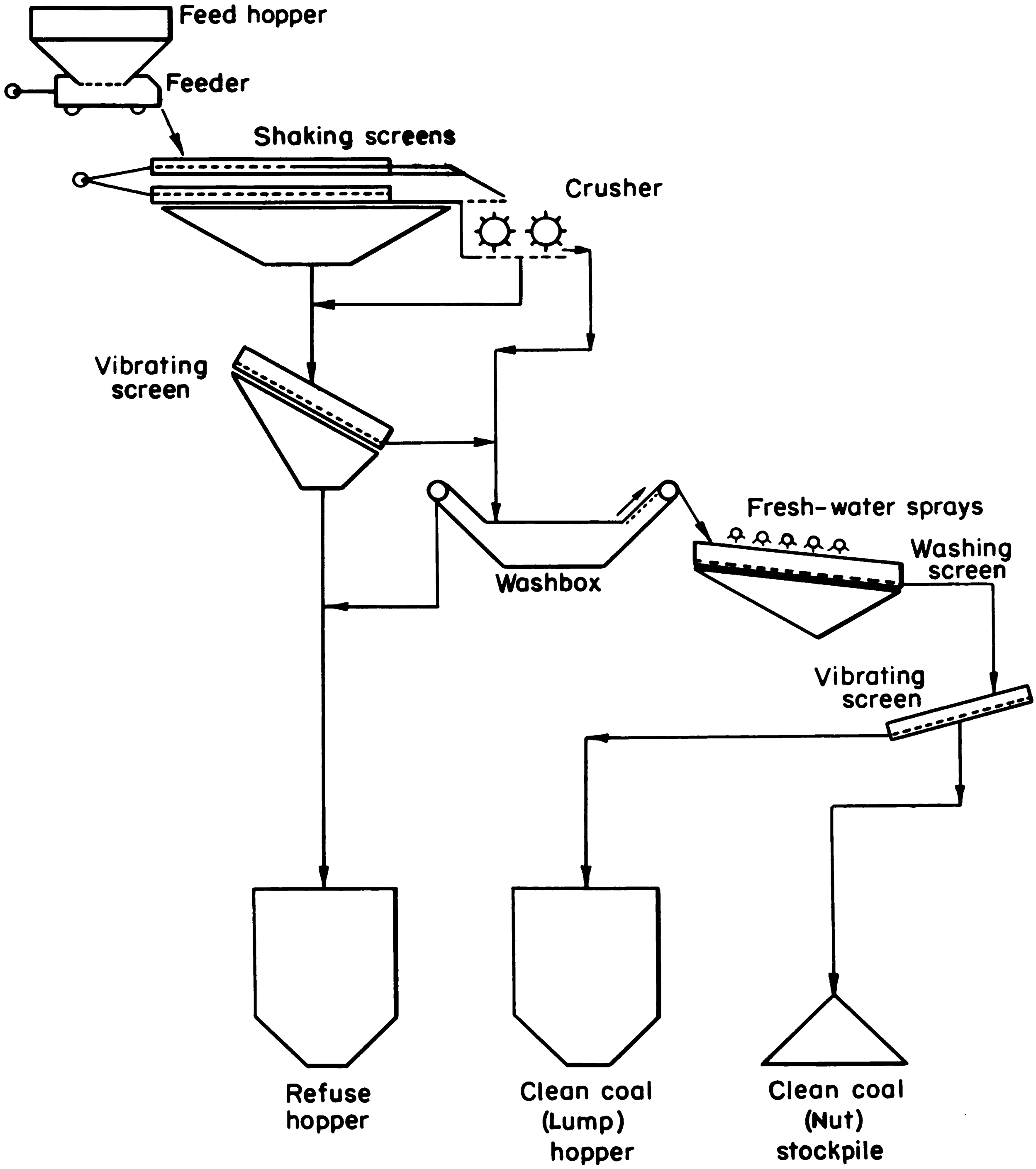

Figure 6. - Flowsheet of washery salvaging coal from Pittsburgh-bed refuse.

The more theoretical phases of this investigation include studies of the chemical and physical interactions between coal surfaces and various chemical agents, factors influencing flotation rates, and the mechanisms of froth- and kerosine-flotation processes. Inasmuch as these studies were initiated during the year, progress has been limited to surveying the literature, laying out a tentative program, constructing and assembling equipment, and training laboratory personnel.

With reference to the more practical phases of the investitation, probably the most serious criticism of the kerosine-flotation process, as used in the Birmingham district, is the relatively small unit capacity of the flotation cells. A study of factors influencing cell capacities was therefore initiated some 2 years ago, and work on this problem is continuing. Early efforts along these lines included studies of the effects of minor changes in cell design and operating procedures on the amount of float coal recovered with a 24- by 24-inch Denver unit cell (rated volume, 12 cu. ft.). Results of these tests indicate that, under certain conditions of operation, significant increases in float-coal recovery, and therefore presumably in cell capacity, may be obtained by installing baffle blocks or similar devices around the impeller diffuser hood at the bottom of the cell.[42] Although it is not claimed that this modification will give increased cell capacities under all possible conditions of operation, the increases noted are considered sufficiently large to warrant additional studies to determine if the conclusions, based on unit cell tests, are applicable to plant practice. Therefore, working in close cooperation with the U. S. Pipe & Foundry Co., a further investigation was undertaken to evaluate the effects of minor changes in cell design and operating procedures on the performance of a battery of four semicommercial cells (Denver No. 15) operated under simulated plant conditions. Present plans include studies of the effectiveness of baffle blocks under varying conditions of operation (now in progress), with a study of the interrelations between feed rate, percentage of solids, recovery, and sharpness of separation. The extent of aeration of the pulp may also be considered, since unit cell tests have indicated that the kerosine-flotation process is highly sensitive to this variable.

Feldspar Jig

At the Northwest Experiment Station another of the Bureau's investigations directed toward improved means of cleaning fine coal is a continuing study of the feldspar jig employed so widely in Europe. Extensive cleaning trials have been made in a laboratory-size, single-compartment unit. With one coal that was moderately difficult to treat, the jig duplicated the cleaning results obtained on a full-size wet table at a unit space capacity 10 times greater than that of the table. Thus, the higher capacity of the jig offers the possibility of substantial economies in washery construction.

Viscosity of Dense Mediums

The dense-medium process is being adopted at a pace outstripping its own technology, and comparatively little is known about the properties of dense mediums. The first phase of a fundamental study of dense mediums now underway is concerned with viscosity. Information on the role of viscosity in dense-medium cleaning may well lead to improvements in separating-vessel design and cleaning-plant operation.

Viscometers suitable for use with unstable suspensions, such as heavy mediums, first were developed and now are being used in evaluating pilot-plant cleaning trials in which mediums with various degrees of contamination are employed (see fig. 7). With some coals comparatively little increase in the viscosity of the medium through contamination with clay impairs cleaning results appreciably. With others, medium or higher viscosity can be tolerated. The investigation has disclosed that the influence of individual types of clay on viscosity varies greatly; therefore mineralogical composition is being correlated with viscosity effects.

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[42] Gandrud, B. W., and Riley, H. L., Increasing Coal Flotation-Cell Capacities: Min. Eng., vol. 7, No. 12, 1955, pp. 1137-1142.

Dewatering Coal Slurry

More stringent enforcement of stream-pollution laws in most of the coal-producing States is necessitating greater attention to recovery of even the finest solids from washery water. In an investigation launched this year, the Bureau is applying vibrating slurry screens, cyclones, vacuum filters, and flocculation to the problem of liquid-solids separation.

Tests in which a slurry screen dressed with 1/4-mm. cloth was supplemented by the action of a cyclone operating on the screen undersize have demonstrated that this combination of equipment can effectively recover virtually all solids coarser than 200-mesh. The high capacity obtained makes the combination economically attractive.

Preparation Characteristics of Coking Coal

As a supplement to the investigation of coking-coal reserves, a program was launched in 1950 to determine the extent to which marginal coals can be upgraded for metallurgical use. Reduction of sulfur generally is the difficult problem, and the experimental work is intended to differentiate among coals for which ordinary washing techniques are adequate, those that require special treatment, and those that cannot be beneficiated adequately with presently known processes. Reports for 1 county in West Virginia and 2 in Kentucky were published during the year, bringing the total to 21.

Wyoming County, W. Va.

The principal reserves of low-ash and low-sulfur coals in Wyoming County are in the Pocahontas No. 3, Beckley, and Sewell beds.[43] Other beds of minor importance as a source of metallurgical fuel are the Pocahontas No. 4, Pocahontas No. 6, Eagle, and Cedar Grove.

The Pocahontas No. 3, Pocahontas No. 6, and Beckley beds are premium low-volatile coking coals and are generally suitable for metallurgical use in the raw, unwashed state. Where the raw coal becomes high in ash, owing to the presence of partings in the bed, a premium product can be prepared in a simple washing operation. The Sewell bed is a low-ash, low-sulfur, medium- or low-volatile coal as mined. The coals from the Eagle and No. 2 Gas beds are of medium-volatile rank and contain moderate percentages of ash and low percentages of sulfur in the raw state. Simple mechanical treatment of the coal from these beds will yield a low-ash, low-sulfur, clean product chemically suited for metallurgical use. The coals from the Cedar Grove, Alma, and Chilton beds are of high-volatile rank, with medium ash and low sulfur content. By cleaning, a low-ash, low-sulfur coal chemically well suited for metallurgical use can be produced.

Studies of the effect of fine crushing upon the release of impurities show that the coals of Wyoming County do not respond to this method of treatment.

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[43] Miller, J. W., Jolley, T. R., and Sokaski, M., Preparation Characteristics of Coal from Wyoming County, W. Va.: Bureau of Mines Rept. of Investigations 5112, 1955, 26 pp.

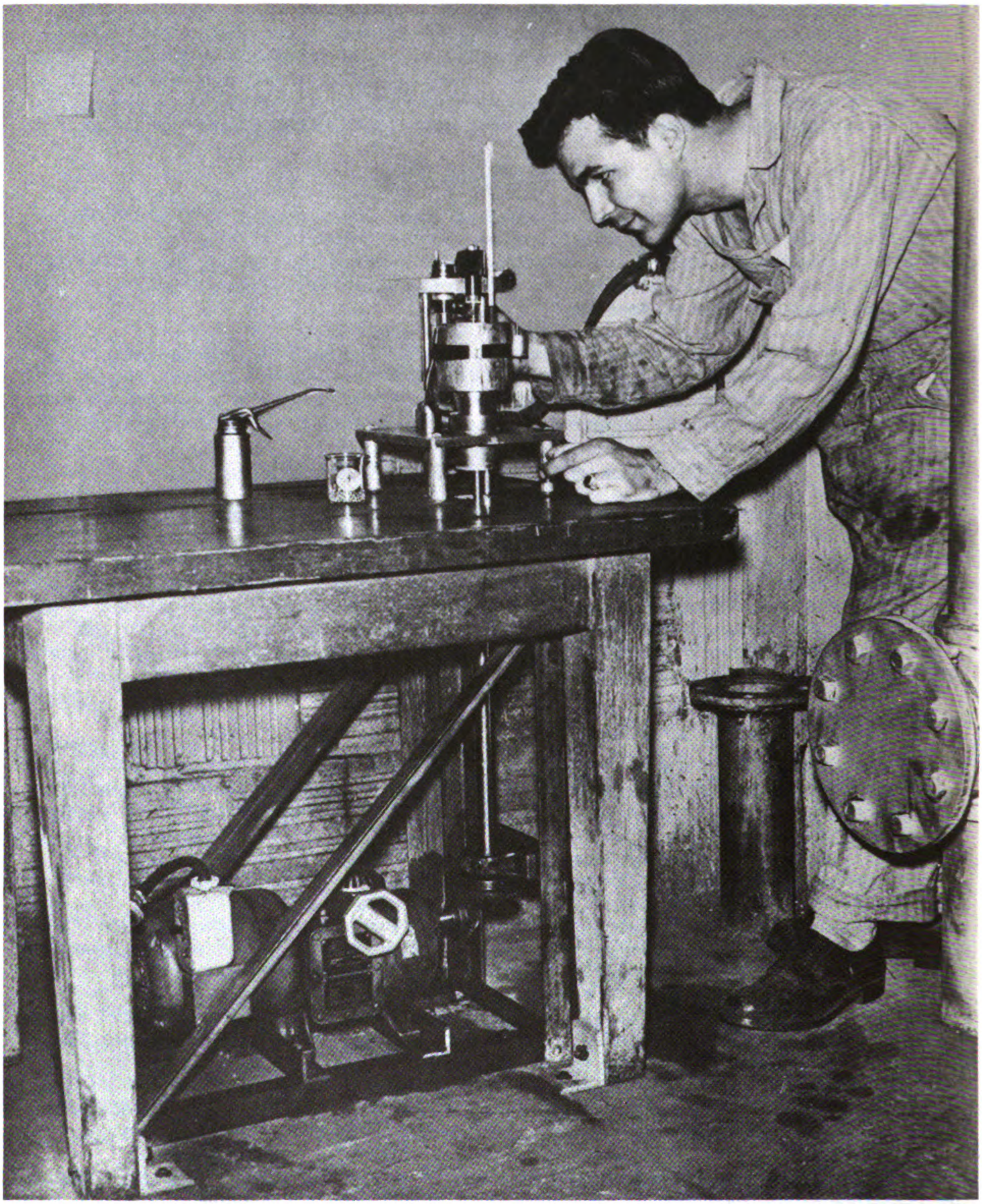

Figure 7. - Stormer viscometer provided with a stirring mechanism to permit testing unstable suspensions.

Letcher County, Ky.

Letcher County coals are of high-volatile rank but, when blended with low-volatile coals, are suitable for manufacturing metallurgical coke.[44] Approximately 31 percent of the coal produced in Letcher County was washed in 4 preparation plants in 1952.

Samples were taken at 19 mines in the Francis, Flag, Fire Clay, Whitesburg, Amburgy, Upper Elkhorn No. 3, Elkhorn Leader, Upper Elkhorn No. 2, and Lower Elkhorn beds for float-and-sink and other analytical testing.

Coal suitable for metallurgical fuel is found in the Francis, Haddix, Elkhorn Leader, and Upper Elkhorn No. 2 beds. The sulfur was low in the coals samples in Letcher County, except in the Amburgy and Whitesburg beds. It was found that in all but one coal sample tested mechanical cleaning would reduce the ash content to acceptable limits for metallurgical use.

Harlan County, Ky.

The coal in the Hindman and Flag beds in Harlan County contains moderate amounts of ash.[45] It is easy to treat coal from these beds to yield a low-ash and low-sulfur metallurgical fuel. If the one sample collected in the Haddix bed is representative of the quality in the reserve area, this bed shows little possibility of becoming a source of metallurgical fuel because of excessive sulfur in the clean coal. From analyses of the samples taken in the Fire Clay and Upper Elkhorn No. 3, these coals appear to be suitable for metallurgical use in the raw state. To upgrade the Amburgy bed to metallurgical quality, fine crushing and precise control will be needed in preparation to reduce the sulfur. Both the D bed and the Lower Split of the Harlan bed contain moderate amounts of ash and low percentages of sulfur in the raw state. Mechanical treatment of these beds to yield a low-ash, low-sulfur clean coal that is chemically well suited for metallurgical use would be easy. The Upper Elkhorn No. 2 bed is believed to be satisfactory for metallurgical use over a wide area, but a high-sulfur area occurs in the central portion of Harlan County near Kenvir, and the bed at this locality is difficult to prepare for use as a metallurgical fuel. The Harlan bed is known to yield a satisfactory metallurgical-grade washed coal throughout most of Harlan County. A high-sulfur area occurs in this bed on Cranks Creek in the central part of the county.

Studies of the effect of fine crushing upon the release of impurities show that the Harlan County coals are unresponsive to this method of product improvement.

A washability study was made of coal from the Pratt bed in the Graysville area of Jefferson County, Ala.[46] These data show that Pratt-bed coal, represented by this sample, when cleaned with conventional jigs and wet-table methods should yield a washed coal analyzing about 5.5 percent ash and approximately 2.0 percent sulfur.

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[44] Miller, J. W., Jolley, T. R., and Sokaski, M., Preparation Characteristics of Coal From Letcher County, Ky.: Bureau of Mines Rept. of Investigations 5135, 1955, 43 pp.

[45] Miller, J. W., Jolley, T. R., and Sokaski, M., Preparation Characteristics of Coal From Harlan County, Ky.: Bureau of Mines Rept. of Investigations 5140, 1955, 44 pp.

[46] Gandrud, B. W., Riley, H. L., Sutton, Paul, Washability Study of the Pratt Bed at the Davidson Mine, Graysville, Ala.: Bureau of Mines Rept. of Investigations 5128, 1955, 21 pp.

In this area Pratt-bed coal has suitable coking characteristics when blended with other coals to make metallurgical coke. The extent of its use is severely limited, owing to its high sulfur content.

Lignite Preparation

The properties and characteristics of lignite differ from those of higher rank coals, and little basic information relating to its preparation is available. Emphasis in research is given to those phases of preparation currently practiced in industry; thereby information is contributed to industrial application and fundamental knowledge of lignite and its properties.

The problems and methods of lignite preparation have been discussed.[47/] Through the years, preparation of lignite has progressed from direct marketing of as-mined lignite to production of a variety of closely sized products involving size reduction, separation by screening, and conveying and handling. Sizes ranging from plus-8-inch lump to double-screened 1-1/2- by 1/2-inch are produced for domestic consumers, and 2-inch by 0 fuel is available for industrial users. A variety of screening and conveying equipment is employed at the mines; however, the toothed double-roll crusher has been adopted almost universally for size reduction of the as-mined lignite, which may be in slablike lumps several feet long. Treatment of prepared sizes consists of spraying with oil to reduce dust in domestic fuel, which is largely shipped in boxcars, and to reduce agglomeration by freezing of industrial fuel, which is shipped in open cars. Consumers utilize the lignite as received, except for two electric-utility plants equipped with pulverized-firing systems.

Because domestic sizes slack during summer storage and mines operate during the winter even though air temperatures may be -30° F., lignite is seldom stored except by industrial users in emergency stockpiles.

Grindability of Lignites

In the laboratory the grindability testing of lignites was studied with the Hardgrove grindability machine.[48/] Owing to loss of moisture from the sample, results by the standard method indicate a grindability index that is apparently too high. To compensate for moisture loss, a "corrected" grindability index was obtained by actually determining the weight of the material that had passed the test sieve. Results of experiments indicated variation of grindability of lignite with moisture content. Minimum grindability was found at midmoisture contents of 23 to 28 percent. Minimum corrected grindabilities of 7 lignites ranged from a low value of 26 to a maximum value of 58. Maximum corrected grindability varied from 41 to 87. For 4 of the 7 lignites tested, the maximum value occurred in the low moisture range at 4 to 9 percent moisture content.

An experimental impact pulverizer, made available by the chemical-engineering department of the University of North Dakota, has been erected in the pilot plant of the station to continue the study of pulverizing characteristics of North Dakota lignites under closely controlled conditions. Supplementing the laboratory-grindability studies, field tests were made on a commercial air-swept bowl-mill pulverizer,

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[47/] Ellman, R. C., Current Problems of Preparation of Lignite Coals: Proc. Coal Conference at Missouri School of Mines and Metallurgy, Dec. 3 and 4, 1953, Univ. of Missouri School of Mines and Metallurgy, Tech. Series 85, pp. 76-86.

[48/] Ellman, R. C., and Belter, J. W., Grindability Testing of Lignites: Bureau of Mines Rept. of Investigations 5167, 1955, 38 pp.

1 of 6 installed at the powerplant of the Central Power Electric Cooperative, Voltaire, N. Dak. A report on these tests was presented at a technical meeting and has been submitted for publication in a technical journal.

Freezeproofing Lignite

Difficulties in unloading are encountered during winter shipment owing to freezing of lignite in the cars. Oil treatment, at the rate of 3 to 5 quarts per ton, has been found effective in reducing the severity of agglomeration. A refrigerated test chamber has been constructed to study the mechanism of agglomeration by freezing, treatments that are effective in reducing it, and optimum application of such treatments. It has been established that agglomeration by freezing occurs by migration of moisture as a vapor from regions of higher temperature at the center of the cars to those of lower temperature near car sides, bottom, and top, where condensation and formation of ice crystals or frost occur. The degree of agglomeration apparently is influenced by the size and number of the crystals formed on the surface of the lignite lumps. Conditions under which the largest crystals develop cause the most severe agglomeration. Preliminary observations indicate that oil sprayed on the surface of the particles influences the crystallizing process, in that numerous hairlike, fragile ice particles are formed, producing weak agglomeration.

Drying Lignite by the Fleissner Process

To provide a relatively stable form of dried lump lignite, lignites from Mercer County, N. Dak., were dried by the Fleissner process in saturated steam at 200, 300, and 370 p.s.i.a. pressure. Net consumption of steam per pound of water removed from the lignite was found to vary inversely with processing pressure and temperature and with source of lignite, and to range between 0.79 and 1.27 pounds of steam per pound of water removed, within the range of pressures and temperatures studied. Data from these studies are compared in a Bureau publication[49] with previous studies of drying North Dakota and Austrian lignites and Australian brown coal.

Lignite Storage

Owing to its high reactivity lignite may heat spontaneously upon exposure to air. During construction of Garrison Dam, Riverdale, N. Dak., rather than waste or market the huge quantities of excavated lignite, the United States Army Corps of Engineers, with the technical assistance of the Bureau of Mines, developed a method of safely stockpiling lignite that has been previously described. To date, over 2 million tons has been placed in storage in 8 piles. Periodically, observations are made on the piles, and temperatures were measured and gas samples obtained at established stations on pile 5. It has been found that the interior of the pile remains stable; however, temperature and gas composition near the pile surface fluctuate owing to seasonal temperature changes and some infiltration of air. The heating value of the stored lignite on a moisture- and ash-free basis has decreased 1.0 percent during a period of 2 years. The potential difficulty in long-term storage of lignite appears to be erosion of the pile by wind and rain. In January 1955 heating developed in a wind-deposited slack drift on one side of pile 5. To prevent heating from spreading to the main body of the pile the slack was moved to the base of the pile, where it cooled. In an effort to control future deposition of the slack, snow fences were installed in three parallel rows on pile 5. Periodic maintenance will be required, however, to remove or level accumulated drifts at these fences.

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[49] Oppelt, W. H., Ellman, R. C., Ongstad, O. C., and Kube, W. R., Experiments in Fleissner Drying of North Dakota Lignite: Bureau of Mines Rept. of Investigations 5122, 1955, 24 pp.

UTILIZATION OF COAL AND OTHER FUELS FOR COMBUSTION

In the foreseeable future the large-tonnage market for coal will continue to be for steam raising and space heating. Technologic advances through the years have greatly improved the efficiency with which coal can be burned, thus husbanding our resources and, at the same time, strengthening coal's position in the highly competitive energy market. Technologic improvement must continue; thus it is appropriate that the Bureau's research should contribute to fundamental knowledge on combustion. The efforts of Bureau technologists in increasing the effectiveness and economy of Government-owned fuel-burning plants is equally important.

Anthracite as Fuel in Frost-Control Heaters

At the request of anthracite-industry representatives the Bureau of Mines has published the results of a technical and economic investigation of the prospects for using anthracite as heater fuel in controlling frost damage to fruit, vegetable, and nursery crops.[50] The study showed that this market for anthracite was not potentially large.

Factors That Affect Heat Transfer in Furnaces

Owing to lack of knowledge of complex heat- and mass-transfer processes that occur within a combustion chamber, furnace design still is highly empirical. It is highly improbable, however, that substantial improvements in dependability and performance or reduction in capital and operating costs will be made until there is better understanding of the nature of combustion and heat-transfer processes in flames and fuel beds and the effects of these processes on the net heat transfer within the furnace cavity. In cooperation with the Special Research Committee on Furnace-Performance Factors of the American Society of Mechanical Engineers, the Bureau of Mines analyzed the data from five boiler stations to determine whether any of a number of correlations that have been proposed by other workers in this field could be used to describe these data. Although the analysis is not yet complete, some significant limitations of the data have been found. A progress report on this work was presented before the American Society of Mechanical Engineers at its Annual Meeting in Chicago on November 11, 1955.

To evaluate the heat transfer in a furnace properly, flame temperatures must be known. The present work is being directed toward this objective. By using the Remington-Rand Univac-60R electronic computer, a series of computations is being made that will indicate the composition of the gas formed in the combustion of coal with air. The limits of the system are determined by the ratio of carbon to oxygen, hydrogen to oxygen, and nitrogen to oxygen. From these data it will be possible to calculate the flame temperatures of various coals. The information obtained at the conclusion of this work will be published.

Fluid Mechanics

Wherever fluids are handled, either for purpose of transfer or of reacting with other materials, as in combustion and chemical processes, increased knowledge of the basic engineering relationships involved aid greatly in the design of efficient equipment. Two aspects of the subject that are being investigated by the Bureau of Mines include complex flow patterns through combustion equipment and the particle-fluid relationships involved in pneumatic transportation of fine powders, such as pulverized coal, and in dust-separating equipment.

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[50] Clendenin, J. D., Frost-Control Practices and the Prospective Use of Anthracite: Bureau of Mines Inf. Circ. 7714, 1955, 45 pp.

The flow-pattern work has been initiated with a literature survey, part of which has been incorporated into an annual review of the literature of fluid mechanics published by a national chemical journal.[51]

The fluid-particle work has included brief studies of the size of fly-ash particles that were made for the Tennessee Valley Authority when that organization was investigating the efficiency of new powerplant equipment.

In cooperation with the Army Chemical Corps, an investigation was conducted into pneumatic transportation of fine powders and the dispersion of powders from small reservoirs. Because of the nature of this work, the results cannot be published in their present form; but, after security clearance, the final reports will be modified to make portions suitable for publication.

In conjunction with the work on fine particles, a novel method of drilling small holes through thick metal was developed and published.[52]

Tests of Army Stoves and Cookers

In cooperation with the Office of the Quartermaster General, a study of the performance characteristics of Army heating equipment was continued.

Endurance tests were conducted on two prototype immersion heaters of similar design, using leaded gasoline as fuel in one and fuel oil in the other. Small carbon deposits were noted in both heaters, on the side of the flue outlet below the baffle plates, after approximately 100 hours of operation. However, continued operation had little effect on the extent of the deposits. Although only slight soot deposits appeared on the draft diverter of the unit using leaded gasoline as fuel, a substantial amount was formed on the diverter of the heater burning fuel oil. When fuel oil is used, frequent cleaning is essential to prevent the buildup of carbon deposits.

When a 4-inch-diameter stack was substituted for the usual 5-inch stack, satisfactory operation was obtained when leaded gasoline was used as fuel, but neither heater would function within the acceptable smoke limit when fuel oil was used.

To facilitate the estimation of thermal efficiency for the different heating units, a chart was prepared showing the percentage of thermal efficiency as a function of carbon dioxide content and temperature of stack gases for diesel fuel oil.

Survey of Industrial Combustion of Lignite in Boiler Plants

Most of the lignite produced in the north central United States is used as fuel for industrial steam-generating plants. Figure 8 shows the location of 21 lignite-burning electric-utility powerplants in North Dakota, South Dakota, and Minnesota and 3 in Canada; the figure also shows mines producing over 100,000 tons of lignite

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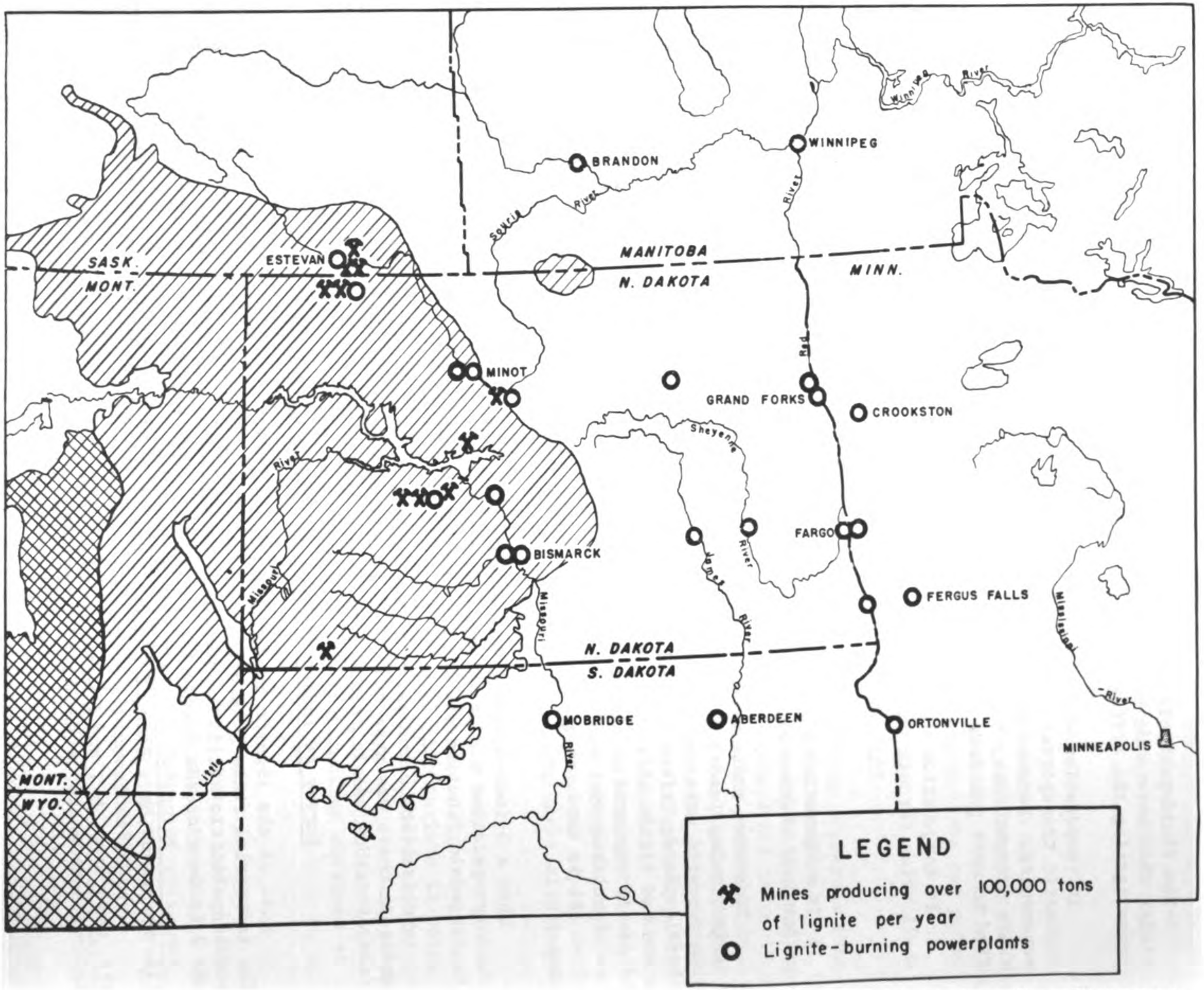
[51] Weintraub, M., Flow of Fluids: Ind. Eng. Chem., vol. 47, part II, 1955, pp. 558-565.

[52] Weintraub, M., and Kollar, G. R., Small-Diameter Holes in Metal: Rev. Sci. Instruments, vol. 26, 1955, pp. 986-987.



of lignite-burning electric-utility plants and principal lignite mines of the Northern Great Plains area.

per year. In view of the importance of this phase of lignite utilization, a survey investigation of large-scale combustion installations now in operation was begun. The purpose of the survey is to obtain direct information on firing methods, performance characteristics, and operational problems at each of the larger lignite-fired boiler plants. Complete specifications are also collected for each individual boiler furnace when available.

Upon completion of the survey, the data will be correlated, evaluated, and published as reference information for those concerned with industrial combustion of lignite. In addition, the survey should reveal recurring problems associated with large-scale lignite combustion that can be used as bases for planning future combustion research.

Twenty-five lignite-burning steam-generating plants, in North Dakota, South Dakota, and Minnesota are included in the survey. Twenty-one of these plants are operated as electric-utility plants, 3 supply chiefly process steam, and 1 is a large central heating plant.

Three different firing methods - traveling grate, spreader stoker, and unit pulverizer - are now in use for industrial firing of lignite. Most of the newer units are spreader-stoker-fired, but there are a few unit-pulverizer installations.

The largest lignite-burning powerplant operated in the area - the W. J. Neal station of the Central Electric Power Cooperative, Voltaire, N. Dak. - has a rated capacity of 30,000 and a maximum capability of 45,000 kw. The newest lignite-burning installation is the 25,000-kw. R. M. Heskett station of the Montana-Dakota Utilities Co., Mandan, N. Dak., shown in figure 9. This plant is unique in North Dakota in that semioutdoor construction is used. Operation of the station has shown this type of construction to be feasible despite severe winter weather.

Fuel-Engineering Service

The Washington office maintains a record of tipple analyses, so that telephonic inquiries can be answered. This service is available for direct contact with Government departments in the Washington area. Analyses are contained in a public file that can be consulted by anyone visiting the Washington office.

Fuel-economy consulting services were given the District of Columbia Sewage Treatment Plant and an Army installation in Maryland. At the latter plant it was found that the induced-draft fan and fittings to the breaching were inadequate to carry the designed maximum load.

A meeting was held between Bureau personnel and fuel-purchasing agents of various Government departments to discuss coke specifications. The primary purpose of this meeting was to review Federal Specification Q-C-571b, and the secondary purpose was to discuss fuel-purchasing problems in general and the desirability of occasional meetings such as those held by the Federal Fuel-Purchasing Committee. Another meeting of Bureau and other Government personnel was devoted to discussion of suggested procedures to be followed by Federal agencies with regard to fuel-inspection work.

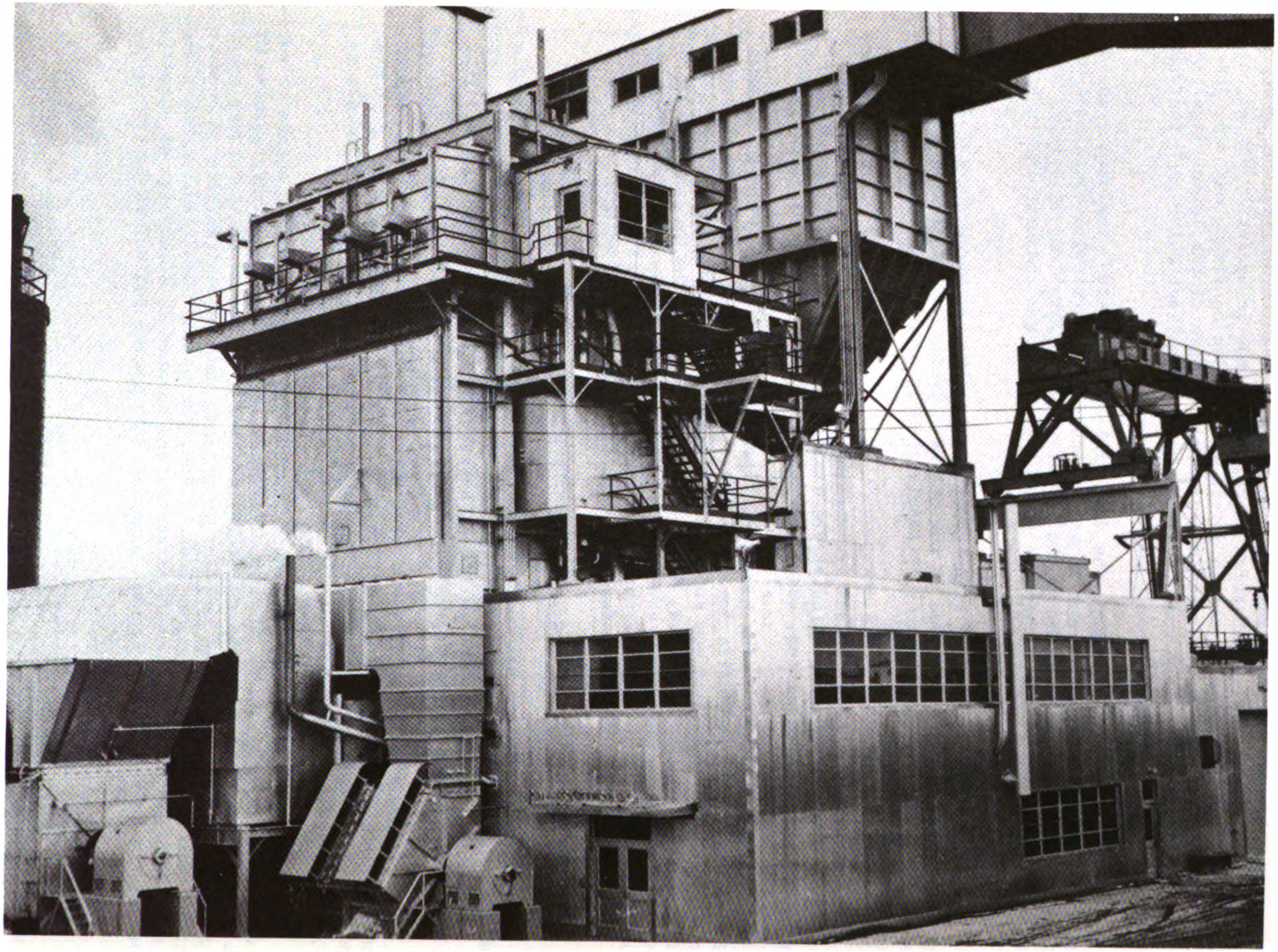


Figure 9. - Semioutdoor, 250,000-lb.-per.-hr. spreader-stoker-fired boiler at R. M. Heskett station, Montana-Dakota Utilities, Mandan, N. Dak.

Original from THE OHIO STATE UNIVERSITY

More detailed instructions on use of the Ringelmann smoke chart were issued,[53] and the results of a series of comparison tests of the Ringelmann chart and a smokescope were published.[54]

OTHER INVESTIGATIONS RELATED TO FUEL UTILIZATION

Use of Anthracite as Metallurgical Fuel

Anthracite as Foundry-Cupola Fuel

As part of the Bureau's program on developing the use of anthracite as metallurgical fuel, melting tests have been run in a commercial-size cupola in which raw and calcined, hard-type, Egg-size anthracite were compared with the foundry coke normally used. Despite efforts to establish as nearly comparable conditions as possible, both the raw and calcined anthracites were less satisfactory than the coke, and desired performance could not be maintained, even for a 4-hour period.

The calcined anthracite was produced by heating raw, Egg-size anthracite to 1,750° F. in a vertical slot oven at average heating rates of 5° to 7° F. per minute. The strength and abrasion resistance of the raw and calcined anthracites, as determined in the ASTM tumbler test on 3- by 2-inch (square-mesh) size, were inferior to those shown by the coke. The stability factor (cumulative percentage on 1-inch, after 1,400 revolutions in the ASTM tumbler) of the raw anthracite was 7 to 15, the calcined anthracite 17 to 19, and the coke 38 to 41.

On the basis of the results from other similar studies, it was concluded that direct use of anthracite in present-day cupolas is not especially promising when high-capacity performance is a deciding factor; however, results obtained during blast-furnace tests at the Bureau's Pittsburgh Station with raw coal tend to alter this viewpoint. These tests, described later, do not indicate enough fuel-bed and stock-column resistance to account for the operating difficulties usually ascribed to the use of anthracite. Samples of fuel that passed through the bed unburned do not show appreciable size degradation when compared with the original fuel charge; and resistance to impact and abrasion, as indicated by the tumbler test, has not been materially altered.

Tests at Pennsylvania State University with a 10-inch cupola using both raw and calcined anthracite showed considerably better size stability in the "drop" material at the end of each test when using calcined coal. These data (confidential) are not substantiated by lower windbox pressures and higher melting rates, but leakage from the cupola taphole may account for this lack of verification.

As it was known that at least 3 large producing companies have been, within the last 5 years, interested in producing calcined anthracite for pot liners for the aluminum industry, as well as for other metallurgical use, the calcination problem was investigated still further. This investigation has resulted in a preliminary agreement to calcine either 1 or 2 carloads of high-volatile, low-ash anthracite at a pilot plant in Pampa, Tex. It is calculated that this plant will be able to calcine 600 pounds of anthracite per hour at 2,000° F. Heating rates, volatile release rates, mechanical size stability, heating size stability, and approximate costs are expected to be predictable from these tests. Enough calcined anthracite will be produced for more extensive cupola tests and for various other fuel and nonfuel investigations.

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[53] Kudlich, Rudolf (rev. by L. R. Burdick), Ringelmann Smoke Chart (ref. of I. C. 6888): Bureau of Mines Inf. Circ. 7718, 1955, 3 pp.

[54] Burdick, L. R., A Comparison of the Smokescope and the Ringelmann Chart: Bureau of Mines Rept. of Investigations 5162, 1955, 12 pp.

The use of some form of physically reconstituted anthracite, made by briquetting, extrusion, or other suitable method of agglomeration, appeared to be a promising supplementary way of introducing anthracite successfully as cupola fuel. A dense, carbonized, agglomerate fuel that proved equal or superior in strength to premium foundry cokes was made by briquetting calcined anthracite fines with about 12 to 14 percent of coal-tar pitch; however, actual tests in a foundry cupola are necessary to determine whether a fuel can be made that has the requisite combustibility and reactivity characteristics of a satisfactory cupola fuel. Anthracite briquets in the 5- by 4-inch and 4- by 3-inch sizes had stability factors of 48 to 50, compared with 32 to 56 for high-grade foundry cokes of these sizes. Anthracite briquets in the 2- by 1-1/2-inch size had a stability of 42 to 69, compared with 37-1/2 to 55 for good coke and 7 to 9 for raw, hard-type anthracite of this size. Studies are in progress aimed at (1) production of tonnage lots of agglomerate fuel for cupola test purposes and (2) process development to permit production of anthracite agglomerate fuel for which the properties and piece size can be varied according to use requirements.

Previous experience has indicated that high-pressure auger extrusion is the most versatile process for producing experimental or pilot quantities of agglomerate fuel. This type of equipment has the advantage of (1) low initial cost, (2) high but easily controlled capacity, (3) low power requirement, (4) low space requirements, and (5) low maintenance costs. In addition, it is relatively simple mechanically to alter the equipment to produce a large variety of briquet sizes.

Arrangements have been made to borrow a machine temporarily for experimental runs and to familiarize station personnel with operating techniques. This machine has a capacity of about 1/2 ton per hour of 1-inch-diameter briquets. It is quite possible that larger briquets can be produced, but no operating information is available on the larger size product. As this is a production machine, its design is proved, and considerable operating experience has been accumulated with it. Meanwhile, inquiries are being made of manufacturers so that equipment can be purchased for the Anthracite Experiment Station.

It is expected that a calcination or carbonizing operation will be required for the process after the briquets are formed. Hence, studies have been initiated to design a horizontal radiant-heated furnace for this purpose. Furnaces now are predominantly of the vertical-shaft type, and present materials-handling problems with green briquets militate against their use in a continuous process. This furnace will be of material assistance in any further calcination studies that may be required.

Lump Anthracite in an Experimental Blast Furnace

Another step in the Bureau's program on the use of anthracite as metallurgical fuel was taken in 1955 with completion of 2 series of tests in the Bureau's 3-foot-hearth-diameter experimental blast furnace at the Central Experiment Station, Pittsburgh, Pa. The first series[55] was run over a period of 2 weeks in the spring of 1955. The results were gratifying, in that as much as 40 percent Egg-size anthracite replaced coke of 2- by 1-inch size with no apparent difficulty in furnace operation. Operation was somewhat rougher, however, with coke-anthracite than with coke alone, using an ore burden consisting of 80 percent as-received Mesabi ore (containing minus-1/8-inch material) and 20 percent sinter.

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[55] Buehl, R. C., and Royer, M. B., Smelting Ore With Anthracite; Bureau of Mines Experimental Blast Furnace: Bureau of Mines Rept. of Investigations 5165, 1955, 15 pp.

Indications were that, under comparable conditions, furnace performance could not be maintained as easily with coke-anthracite as with coke, which is in general agreement with observations growing out of the successful operation of early anthracite (and coke-anthracite) blast furnaces, as well as recent experience in which anthracite was used with coke in experimental and commercial furnaces producing spiegeleisen and in furnaces producing pig iron and ferromanganese. The metal produced by smelting with anthracite in the first series of tests was low in sulfur (0.013 to 0.059 percent) and of uniform silicon content (0.88 to 1.97 percent), within the normal range. The short time available for these tests did not permit determination of the best production and coke rates. With the lower burden ratio (about 1.5 to 1.6) of iron-bearing materials to fuel used in this series, there was no evidence that a higher ratio could not have been used, which would have increased production and decreased the fuel rate, as was indeed demonstrated in the second series of tests.

In the second series, conducted over a period of 4 weeks in the autumn of 1955, the results with as much as 60 percent anthracite, in both Egg and Stove sizes, were much improved over those in the first series. Iron-bearing materials used in this series consisted of Erie taconite pellets in 1 group of tests and 50-percent Venezuelan ore and 50-percent ore sinter in the other group, with burden ratios of 1.80 to 2.24, and 1.80, respectively. Metal produced in this series was generally comparable, but with the sulfur contents (0.027 to 0.114 percent) running a little lower than in the first series. Metal-production rates were higher and fuel rates lower than those in the first series, in agreement with the prediction from the results of the first series in which it was stated that higher burden ratios should operate in this direction. However, contrary to previous experience, operation with anthracite-coke mixtures was much smoother than during the early tests and was, for all practical purposes, as smooth as with coke alone.

Burden ratios as high as 2.24 were used; however, silicon (0.95) and sulfur (0.073) in the metal were not satisfactory. Acceptable metal was obtained at ratios in the range of 2.0 to 2.11. Fuel rates were decreased to 1,295 pounds of fuel per ton of metal with fairly satisfactory operation. Higher burden ratios and lower fuel rates were maintained with anthracite-coke mixtures than with coke alone as the fuel.

The effect of hot blast temperature on fuel rate and production rate in this series is shown in figure 10. As may be seen in this chart, the better fuel and production rates at the higher burden ratios also coincide with the higher blast temperatures. The significance of the differences in rates between the runs with pellets and with the ore sinter cannot be fully assessed at this time because of the limited operating experience with the latter (153 hours) compared with the former (373-1/2 hours) and results at only 1 burden ratio on the ore-sinter burden. By using higher burden ratios, it appears that production and fuel rates with this burden will be considerably improved and approximately comparable with those for the pellets.

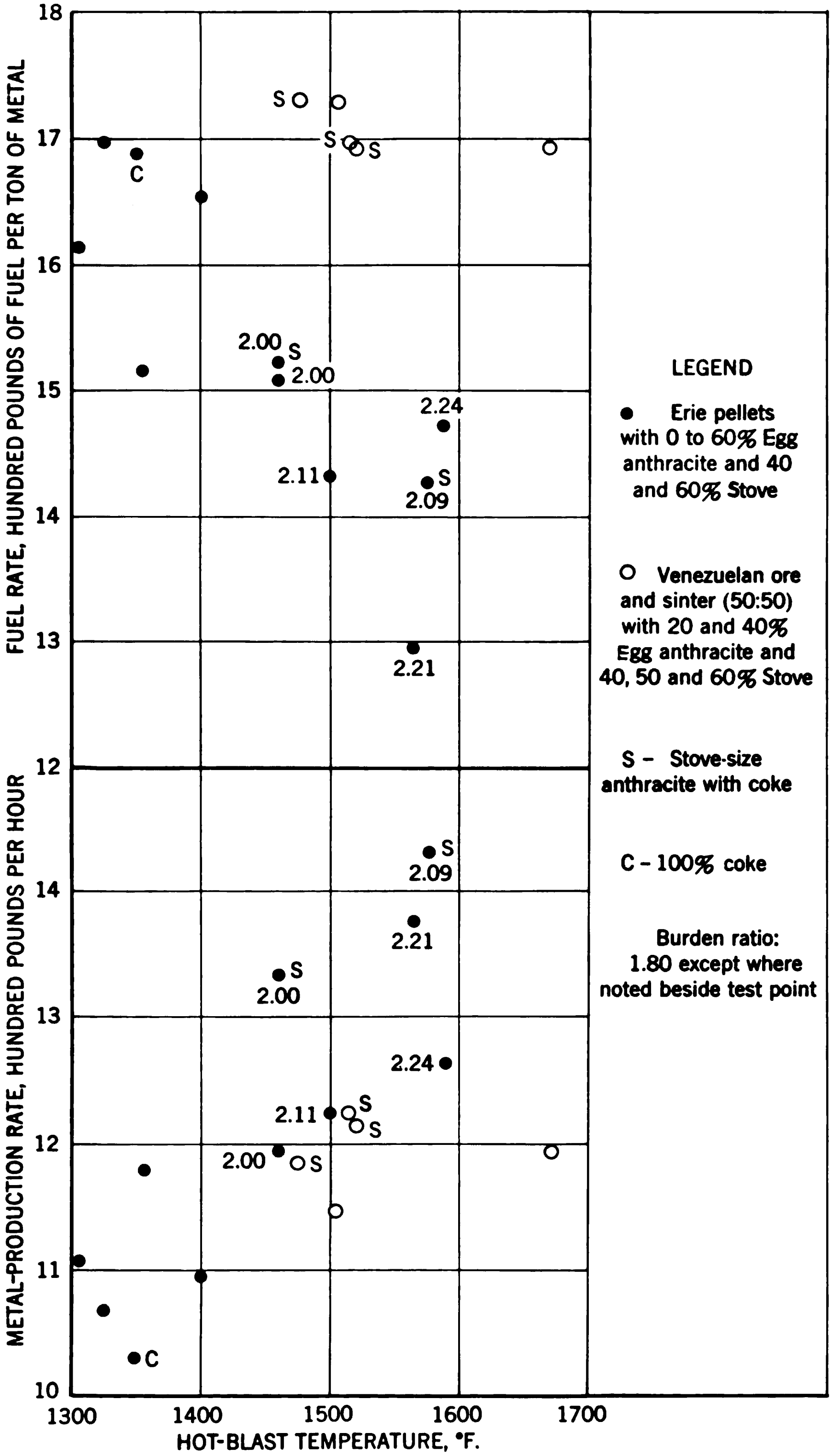

Figure 10. - Effect of hot blast temperature on fuel rate and production rate.

Lump Anthracite in a Blast Cupola

As a still further step in investigating the use of anthracite in metallurgical blast furnaces and remelting cupolas, a cooperative arrangement was made with an eastern Pennsylvania steel plant to observe operation of a commercial-size furnace of this type in which Egg-size, standard-grade anthracite was to be substituted for increasing proportions of the total fuel required. Anthracite-producing companies participated in the project and contributed the coal. The furnace had an internal hearth diameter of 7 feet and a stock-column height of 28 feet above the tuyères. It was designed to work with blast air preheated at 1,000° F. and to discharge molten iron and slag continuously in separate streams. The operations were supervised and conducted by the steel-plant personnel. Operating data and material samples were collected by Bureau staff.

A great variety of low-grade ferrous scrap, mostly steel, was used with limestone flux, silicon additives, and fuel to yield a recarburized, low-sulfur, molten iron for feedstock to the openhearth steel furnaces in the plant, which were producing ingots for low-carbon plate (see fig. 11). Metallurgical coke had been the fuel in the previous short breakin runs of the equipment and was continued as the base fuel in the current tests, during which there were 2 runs of 7 and 6 days, in which anthracite replaced some of the coke. In the second of these, anthracite was started at 20 percent of the total fuel and increased at 2- and 1-day intervals to 35 percent. Operating difficulties, both mechanical breakdowns and trouble in maintaining slag flows, were frequent during the observation period but seemed to be no different with some anthracite in the fuel than with all coke. The output of the furnace averaged over 16 tons of molten iron per hour with the 20- to 25-percent anthracite during the 6 days, and this was more than had been obtained in a base-data coke run. Fuel consumption in the 6 days averaged 3-3/4 pounds per pound of hot iron and was about the same as with all-coke fuel.

These preliminary tests indicated that anthracite may be substituted in some proportions for coke in a metallurgical blast cupola without loss in capacity or increased fuel consumption. The limit in the proportion of anthracite did not appear to have been reached in the time available for the tests. However, the effect of higher percentages can only be established by longer runs, with more freedom from interruptions for mechanical difficulties, so that there is adequate opportunity to adjust the flux materials for a workable slag and to reach equilibrium in the reactions of the two fuels. This process appears particularly favorable for the use of anthracite in vertical metallurgical furnaces, since the weight of the burden in the short stock column is less and the time for working down to the reaction zone is shorter. Also, the highly irregular shape of the pieces of steel and cast-iron scrap help to maintain the permeability of the stock column all the way down to the melting zone without any great dependence on low fuel density.

Anthracite as Reducing Agent in Electric-Furnace Production of Phosphorus

A preliminary investigation of needs and requirements for use of anthracite as reducing agent in the electric-furnace production of phosphorus was conducted in 1955. This disclosed that the announced withdrawal of coke breeze from the open market in 1956 by one large supplier is expected to decrease availability and push the price of coke breeze upward, with the result that anthracite is receiving renewed consideration. Previous commercial-scale studies by one large phosphorus producer demonstrated only limited applicability for raw anthracite because of its poor thermal stability. Since calcination is known to improve this property, work is in progress to determine whether calcined anthracite has enough stability for

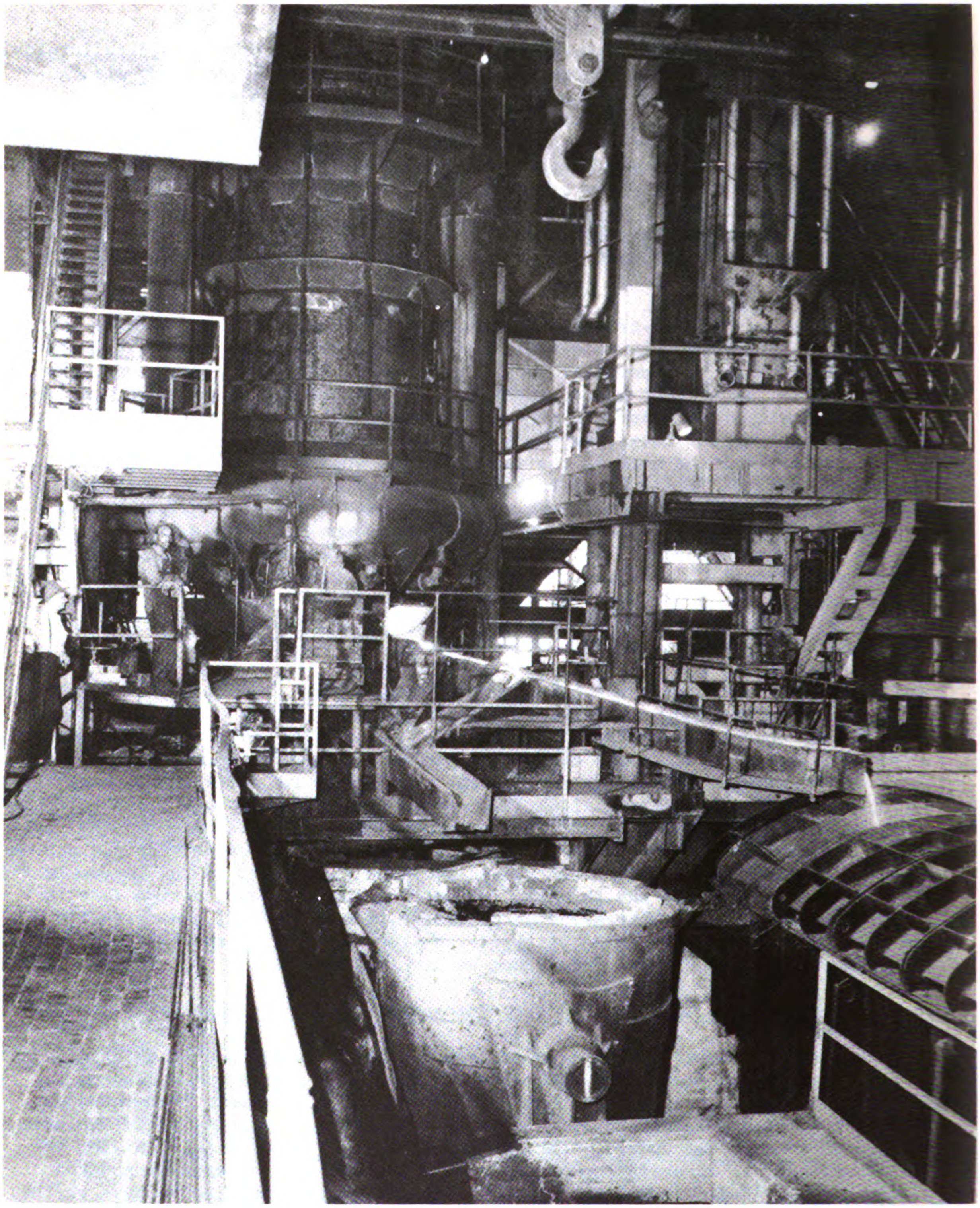

Figure 11. - Metallurgical blast cupola producing molten pig iron, using anthracite for part of the fuel requirements.

this application, which, in the territory that can probably be reached economically from the Pennsylvania anthracite region, indicates a possible annual potential of upward of 100,000 to 125,000 tons.

Germanium From Anthracite

The project, begun in 1954 on recommendation of the Anthracite Advisory Committee, for a survey of flue dusts from anthracite-burning steam-generating plants as a source of germanium, was completed, with negative results. Spectrographic analyses of the flue-dust and coal samples collected from eight plants in the anthracite region were received. Ashes from the coals showed, at most, only "barely detectable" amounts of germanium. The best 3 of the flue-dust samples - all from stoker-fired boilers - contained 100 to 170 parts per million. Under present market conditions, in the face of competitive materials now available, at least 500 parts per million of germanium are said to be necessary for economic recovery.

Hydraulic Transportation of Bituminous Coal and Lignite

The hydraulic transportation of bituminous coal has been studied by private industry and soon will be applied in Ohio. A similar technique might be attractive in the West in locations where individual coal reserves are not sufficient to warrant location of a powerplant. It may prove economically feasible to work out individual deposits, one at a time, by hydraulically transporting the coal to a central powerplant, and a study is being conducted at the Denver laboratory on this problem. The study will cover experimental work on the degradation of lignite in a pilot pipeline, an investigation of dewatering methods, and a general economic evaluation of hydraulic transportation as applied to Texas lignite.

Lignite and coal undergo an unusual type of degradation under conditions of pipeline transport, and this has proved to be a major problem. Size-consist curves for 1/2- by 0-inch and 1/4- by 0-inch feed sizes of Texas lignite and Colorado bituminous coal, that have been moved roughly 75 miles through a 3-11/16-inch-inside-diameter pipeline, are shown in figure 12. Deviations from ordinary behavior are apparent from the fact that whereas, when the Rosin-Rammler system of plotting size distribution is used the curves are linear or nearly so, these curves display curvature resulting from the presence of substantial quantities of ultrafine solids.

Although enhancing pipeline flow, the production of fine material is detrimental because it aggravates the problem of removing the water at the pipeline terminus. The necessity for dewatering and thermal drying, which is independent of the length of the line, makes short lines difficult to justify economically, particularly as applied to lignite.

Properties and Utilization of South Arcot, India, Lignite

Reference has been made earlier in this report to the Bureau's cooperation in the Government's program of technical assistance to undeveloped areas in foreign countries and the request by the International Cooperation Administration for recommendations on the mining, preparation, and utilization of lignite from the large, but as yet undeveloped, lignite reserves in the South Arcot district of India. Development of these deposits and their efficient utilization depends upon thorough understanding of the properties of the lignite and application of past experience in utilizing similar materials. Samples of the lignite have been tested at the Denver laboratory by the techniques developed in studying western United States coals. The drying and carbonizing properties have been evaluated and preliminary studies

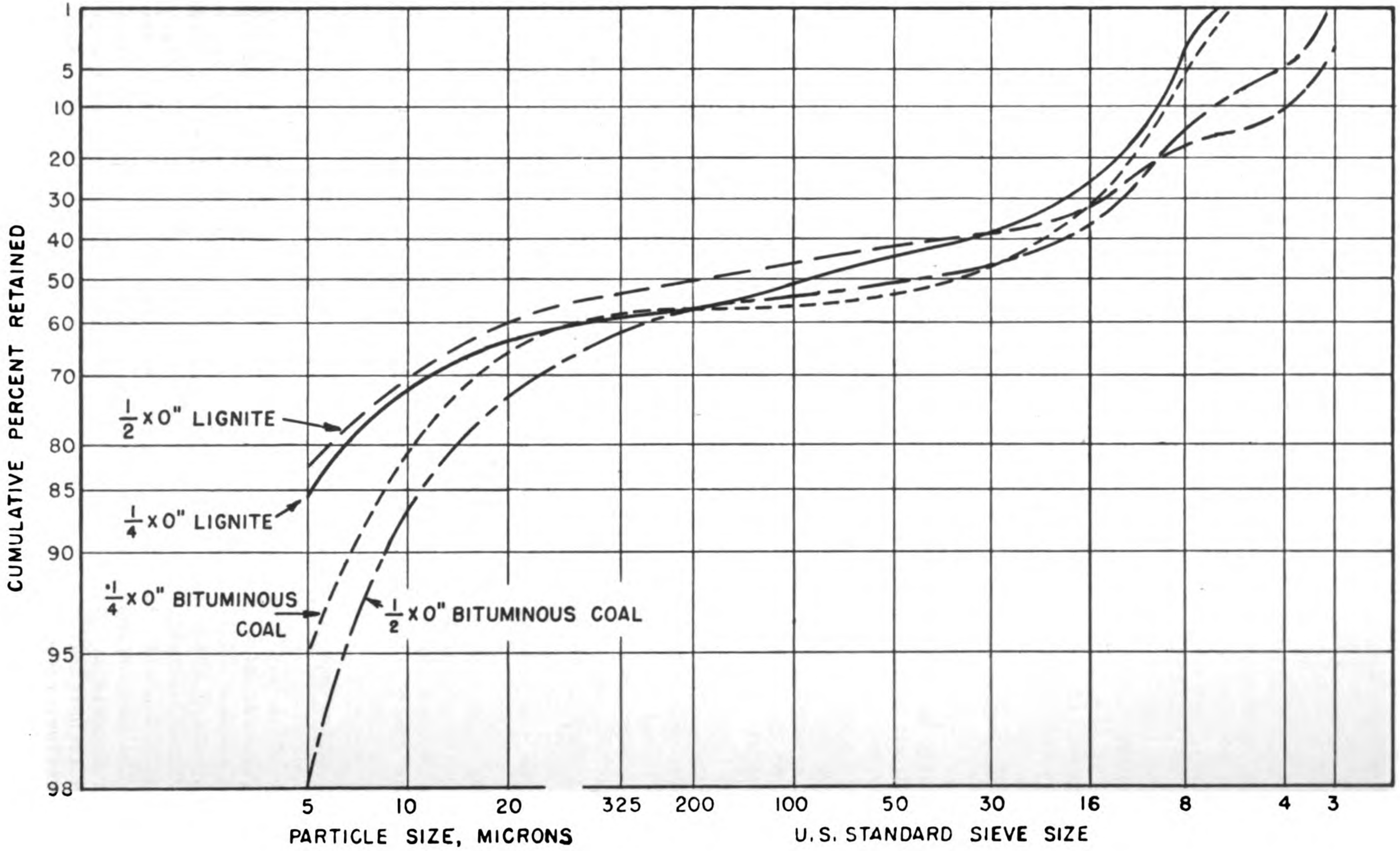

Figure 12. - Size consists of lignite and bituminous coal after 75 miles hydraulic transport.

conducted on binderless briquetting of the partly dried lignite. Based on this experimental work, a workable scheme has been prepared for utilizing Indian lignite.

The South Arcot lignite is similar in chemical properties to the high-moisture lignites of the United States. It is estimated that as mined it has an average moisture content of 56 percent and a heating value of 4,860 B.t.u. per pound. One of the samples dried in the fluidized-pilot-plant unit contained 35.2 percent of moisture as received and had a heating value of 7,220 B.t.u. per pound. It was readily dried to a moisture content of 2.5 percent and a heating value of 10,960 B.t.u. per pound.

On carbonization a ton of the dried lignite produced 1,280 pounds of 12,170-B.t.u. char, 21.5 gallons of tar plus light oil, and 7,850 cubic feet of 180-B.t.u. gas. Both the dried lignite and the low-temperature char are excellent fuels for use in powerplants or cement kilns. Because of its extremely high moisture content, the raw lignite cannot develop a high enough temperature to be used in burning cement, but either the dried lignite or the char should be very satisfactory for such purposes.

It was demonstrated that excellent binderless briquets could be made from the partly dried lignite. Because of their cleanliness, high heating value, and physical stability, such briquets should be attractive industrial or domestic fuel in India. Lump char produced by carbonization of the raw lignite was too soft and friable for industrial use, and the only metallurgical application of this lignite would probably be through carbonization of briquetted dried lignite.

Computations on the drying of the South Arcot lignite showed that a plant capable of processing about 250 tons of raw lignite per hour would produce enough fuel for the requirements of a 200,000-kw. thermal powerplant. A 25-ton-per-hour drying unit would furnish fuel for a cement plant producing 6,000 barrels of clinker daily. Such installations would provide an excellent starting point for industrial expansion based on utilization of the South Arcot lignite.

Boiler-Feed-Water Conditioning

The Industrial Water Laboratory furnishes consulting boiler-water service to Government-operated heating and power plants to improve their operating economy; yearly savings of $32,000,000 are credited to this service.

Recommendations were made on analyses of 13,218 samples of boiler water submitted by Federal agencies in 1955. The bulk of these samples were from the Department of Defense and the Veterans Administration, while 13 other agencies supplied the remainder. Also, 1,119 Bureau of Mines field water-test kits, 27,092 test-kit replacement parts, and 20,533 bottles of chemical reagents were sent to Federal agencies during the year.

A few examples of specific problems on which assistance was given follow: Use of neutralizing and filming amines to control return-line corrosion; preventing furnace-tube failures; return-line corrosion testing to determine corrosion rate; control of condensate contamination; control of boiler sludge deposits; protecting idle boilers from corrosion; and feeding procedure for phosphate and sulfite chemicals. Agencies receiving these consulting services included the Departments of the Air Force, Navy, Army, Justice, and Post Office, the Veterans Administration, Bureau of Indian Affairs, Coast Guard, and the District of Columbia Government.

Boiler-Water Research

Return-Line Corrosion

The cost of replacing return-line piping, corroded by carbonic acid in the condensate, continues to plague Federal heating plants. For example, over $2,500,000 was spent in the first half of 1955 to repair or replace steam-return lines at Air Force installations in the continental United States alone.

About 400 Bureau of Mines condensate-corrosion testers were supplied to Federal heating plants during the calendar year, and approximately one-sixth have been returned for evaluation of the test results. Although the number of heating plants surveyed is a relatively small proportion of the total operated by the Government, the data accumulated indicate that chemical treatment for controlling condensate corrosion is necessary in less than 15 percent of the plants studied. It has also been shown that volatile alkaline amines are practical and dependable for chemical treatment in plants where nearly all the steam is condensed and returned to the boilers. Spectacular reductions in corrosion have been effected by such treatments, frequently at an overall cost of less than $1 per million pounds of steam produced. The survey program is effecting substantial savings to the public, far beyond its costs, both by discontinuing or forestalling expensive treatment costs that are unnecessary and by substituting treatment costs for much larger maintenance costs wherever treatment is warranted.

It is noteworthy that the Bureau's work on the use of alkaline amines to protect steam-condensate piping against corrosion has been largely adopted by industry, especially for the central powerplants in which most of the Nation's electricity is generated. By effecting a substantial reduction of corrosion of the preboiler piping and heater systems, the amine treatment minimizes the quantity of iron and copper that enters the boiler with the feed water. Inasmuch as oxide deposits of these metals have been associated with expensive shutdowns resulting from boiler-tube failure, amine treatment is regarded as one of the more important recent advances in the chemical engineering of boiler-feed-water conditioning. Morpholine has been shown by the Bureau to have especially useful properties for such treatment, but there is some uncertainty as to the stability of this chemical at the very high temperatures and pressures, above the critical point, of the newest steam generators.56/

In connection with the use of hydrazine to scavenge oxygen from boiler water to control corrosion, it has been pointed out that zero dissolved oxygen does not guarantee freedom from corrosion, since steel can be severely oxidized by boiler water under special conditions.57/

Fuel-Oil Additives

Research pertaining to fuel-oil additives was begun to meet requests from many Government agencies for information on alleviating sludge and sooting difficulties experienced in plants burning No. 6 fuel oil. The sludge problem was especially troublesome for a variety of reasons, including lost tank capacity, overloaded pumps, fouled heaters, and clogged piping. Troublesome soot deposits necessitated special cleaning and were thought to promote corrosion and reduce fuel efficiency.

56/ Berk, A. A., Discussion of Experimental Boiler Studies of the Breakdown of Amines: Trans. Am. Soc. Mech. Eng., vol. 77, 1955, p. 452.

57/ Berk, A. A., Discussion of Corrosion of Steel in Boilers: Trans. Am. Soc. Mech. Eng., vol. 77, 1955, p. 441.

It was found that the No. 6 fuel oil, as delivered to storage tanks, contained a very small proportion of solid carbonaceous materials and water that settle out together to form sludge of a quality similar to that found in the tanks. Proprietary additives tested in a laboratory settling apparatus were found to have little or no effect on the sedimentation process. Field surveys of tanks receiving the same oil in the same distribution area disclosed that those in which additives were used had accumulated at least as much sludge as those receiving no treatment.

Chemicals were found that, even in low concentration, delayed the sedimentation of water and of some kinds of carbonaceous material. However, the effectiveness of these chemicals (all in the group of quaternary ammonium compounds in which each hydrogen is substituted by an organic radical) was uncertain, and depended on careful control of relative concentrations and other factors that are not practically achievable in industrial operation. Thus, it could not be recommended that these chemicals be compounded with additives, especially as the present price of such treatment would be high compared with the cost of the oil.

One method of preventing intolerable sludging in storage tanks is what may be called "controlled bunkering."[58] The contribution of sediment from one truckload of oil is small; if it can be prevented from accumulating with sediment from previous truckloads, no great accumulation will result. This hypothesis was tested in a storage tank that was refitted so that oil was bunkered through a pipe that went nearly to the bottom of the tank to stir up any material that had settled out. Oil samples were taken at various levels in the tank before and after bunkering, and the sample quality showed that sludge dispersion was good and reprecipitation was not instantaneous. A low-suction takeoff, about 6 inches off the bottom of the tank, caused much of the sediment to be carried out with the oil to the furnace. The slightly sludged oil burned without difficulty. Considerably less sludge was found in a tank operated 1 year with controlled bunkering than in an adjacent tank that was bunkered through a hatch. Apparently, what was taken out through the suction takeoff when bunkering was controlled corresponded roughly to the sediment added with the new oil. Operators using this scheme of controlled bunkering have not been troubled with sludge.

It is well known that certain chemicals, notably copper and lead salts, can be used to control the accumulation of soot in the flue-gas passages of coal-fired boilers. Oil-soluble forms of such chemicals were used to determine whether or not relatively small concentrations of the chemicals could be added to No. 6 fuel oil to control the accumulation of soot on heat-exchange surfaces. Air-cooled probes were employed for collecting soot in the gas passages of a laboratory furnace burning a mixture of No. 6 and No. 2 fuel oils in the ratio of 1:2 by volume. Surface temperatures of the probes ranged to 300° F., and the flue-gas temperature at the probes was 625° F. or higher. Dosage of additive chemicals was considerably greater than that recommended for the average compound sold for this purpose.

The reproducibility of soot formation was poor in that the weight of accumulated soot was not the same in duplicate tests. Consequently, no numerical values can be given to compare the relative effectiveness of the various chemicals as soot removers. However, the results indicated that none of the additives effected appreciable improvement under the test conditions. Also, flame conditions were not visibly improved by using additives, and the density of soot and smoke in the flue gases was not visibly diminished.

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[58] Barkley, J. F., Hopps, G. L., and Berk, A. A., The Sludge Problem in No. 6 Fuel-Oil Tanks: Bureau of Mines Rept. of Investigations 5113, 1955, 14 pp.

Examination of the probe deposits showed that using metal-organic additives in the fuel increased the total ash content of the soot considerably and that this increase was due to the additive metal, which, in some form, was deposited on the probes as an integral part of the soot. The inability of the fuel-oil additives to effect substantial reduction of soot accumulation on the probes of the test furnace cannot therefore be ascribed to any failure of the additive to reach the soot; instead, it would appear that the temperature was not high enough for the additive to be effective. The results of the investigation thus tend to show that the use of chemical additives to reduce accumulation of soot in low-pressure oil-fired boilers would be ineffective where the metal surfaces, upon which the soot collects, are 300° F. or below.

The experimental use of fuel-oil additives to control deposition in and corrosion of central-station boilers has been followed with interest, especially as to procedures and devices for determining the corrosivity of flue gas. One method depends on analysis of flue-gas samples for SO_2 and SO_3, with a dewpoint meter furnishing corroborating data. Unfortunately, it is very difficult to preserve the quality of the flue gas during sampling, and the results obtained by the method apparently are not of established validity.[59]

Stream Pollution

Research on stream pollution is concerned with procuring quantitative data on waterborne industrial wastes of mineral origin and with developing methods to abate pollution.

Inspection of mineral-industries plants in New York and the New England States has been continued, and a report providing quantitative information on waste-water flow rates and concentrations of mineral materials at 1,025 plants has been prepared for publication. The report identifies the specific origin of waste from various processes, describes novel waste-abatement practices observed, and makes detailed recommendations on steps that can be taken to reduce stream pollution.

A pilot plant and equipment have been designed to be available to interested industrial plants for studying regeneration of the free acid and ferrous sulfate monohydrate from spent pickle liquor. The simplicity of the process employed comes from the laboratory findings that ferrous sulfate readily begins to precipitate when the acid concentration is no greater than 25 percent sulfuric acid by weight and that the crystals will settle out from a 35-percent sulfuric acid solution to leave a clear supernatant liquid after only 30 minutes settling time. The pilot plant would permit determination of the savings in acid, labor, steam, and water that would be achieved by regenerating spent pickle liquor. It may also demonstrate a saving in pickling time and better control of pickling quality. The process is particularly adaptable to plants that have no space available for disposing of the sludge from acid neutralization.

Cooperation with the United States Public Health Service was continued by receiving 11 pollution-control reports on river basins in the United States. Review of these reports produced constructive criticisms that have been incorporated into the final draft of each pollution-control report.

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[59] Berk, A. A., Discussion of the Use of Additives for the Prevention of Low-Temperature Corrosion in Oil-Fired Steam-Generating Units: Trans. ASME, vol. 77, 1955, pp. 275-276.

Informal cooperation with industry was carried out by 14 plant visits and attendance, at conferences, of engineers concerned with stream-pollution problems. Recommendations were made on practical methods and processes to abate waterborne industrial wastes of mineral origin.

Laboratory and pilot-plant research was carried out on removing suspended oil globules from cooling water discharged from the cold rolling of steel. Most of the oil was in globules about 0.0001 inch, in diameter, and the oil remained suspended in the water even after 24 hours settling time. A forthcoming report of investigations on this work will detail the data secured with settling, filtration, aeration, centrifuging, flocculation, and flotation of the cooling water.

Waste Waters From Sand and Gravel Plants

In 1953, 440,089,225 short tons of sand and gravel was produced in the United States. Approximately 400 to 460 gallons of water flows to waste from the washing of each ton of sand and gravel. Because this water contains about 5 percent or more of suspended sand fines and clay, it is estimated that about 40 million tons of material is discarded either to the local streams or as land fill. Such a large tonnage of available waste warrants investigation to ascertain if it contains useful material.

Pilot-plant studies in a liquid-solid cyclone indicate that about 50 percent of this material is salvageable plus-325-mesh sand fines. The principal concern of operators of sand and gravel pits about the loss of all this material is the unsatisfactory public relations caused by its discharge to the local streams. Plant visits to sand and gravel pits are planned to determine the costs for disposing of the waste water and to highlight possible profits from utilizing the discarded material.

Waste Water From Cleaning Diesel Engines

A stream-pollution problem caused by the discharge of waste water from cleaning and maintaining the diesel engines belonging to a railroad was also studied. Recommendations were made that batches of alkaline, oil-bearing solutions be transferred to a holding tank for continuous low-volume discharge and that a quantitative study of the plant wastes be conducted.

The quantitative study showed that the usual oil concentration of about 200 p.p.m. in the plant effluent was increased to over 12,000 whenever the small batches of alkaline solutions were dumped; the pH of the plant effluent also increased from 8.2 to over 10.0. The total flow of waste water was fairly constant at about 140 g.p.m. Therefore, a temporary retention basin was erected at the waste-water outlet to study the possible improvement that would be effected when the pH of the waste water was lowered. Enough dilute hydrochloric acid was added at the basin to bring the pH down to about 7.0, and the oil content of the water discharged from the basin was reduced to about 35 p.p.m.

These results were so satisfactory that the railroad system is now constructing complete retention facilities at a total cost of only $45,000. There will be a concrete retention basin and a spare, each equipped for skimming oil and removing settled solids. Facilities will include pumps for transferring oil and solids and tanks for storage and continuous feeding of acid. It is expected that there will be no significant additional labor cost for this installation, because the same man who is in charge of the raw-water supply for the yard will operate the waste-treatment plant.

COAL CARBONIZATION

Analysis of Coking-Industry Accident Data

Detailed statistical reports on deaths and injuries in the coking industry for 1952[60] and for 1953[61] show sustained progress in the reduction of injuries to employees as compared with 1951. The coke-manufacturing industry established a new safety record in 1953. The combined fatal and nonfatal injury rate of 6.90 per million man-hours of exposure for the industry was the lowest ever recorded, and the number of fatal (8) and nonfatal injuries (425) was lower than in any year since 1939. In 1952, 8 men were killed and 546 others injured. Accidents that resulted in the most injuries involved falls of persons, burns, handling objects, haulage equipment, falling objects, and handtools. Preliminary figures for 1954 show 8 fatal and 275 nonfatal injuries, continuing the favorable trend.

Survey of Carbonizing Properties of American Coals

The program of establishing the carbonizing properties of American coals was continued, using the Bureau of Mines - American Gas Association (BM-AGA) apparatus to determine yields of coke, tar, coal chemicals, and gas. This work is providing industry with an appraisal of the carbonizing properties of coals and blends that is proving even more important as new sources of coking coal are sought to fuel the expanding metallurgical industry. Begun in 1928, the survey has embraced several hundred American and foreign coals; during 1955, 31 additional coals were added to the list. They were from significant reserves in Logan County, W. Va.; Harlan and Bell Counties, Ky.; Campbell County, Tenn.; and Westmoreland, Fayette, Jefferson, and Cambria Counties, Pa. All of these coals were carbonized at 900° C., as were blends of 4 Kentucky coals and 1 Pennsylvania coal. A blend of 4 coals from Formosa was carbonized at 900° and 1,000° C.

West Virginia Coals

The West Virginia coals were from Logan County and are high-volatile A bituminous in rank, with dry, mineral-matter-free fixed carbon ranging from 56.5 to 64.8 percent. Upper Cedar Grove coals from 2 mines and Stockton from 1 mine contained over 10 percent ash; No. 2 Gas coal from one mine and Alma from another contained 2.0 percent or more sulfur; they are therefore undesirable as metallurgical coking coals unless their chemical properties could be improved by cleaning. Lower Cedar Grove coal samples from 2 mines, 2 samples of No. 5 Block, and Buffalo Creek coal from a prospect opening were chemically suitable for metallurgical use and yielded cokes with strength indexes about normal for their rank.

Kentucky Coals

Harlan County. - Flag-, Fire Clay-, Upper Elkhorn No. 2-, and Upper Elkhorn No. 3-bed coals were of high-volatile A bituminous rank and contained less than 0.9 percent sulfur and 4.5 percent ash. These coals differed significantly in cokemaking properties when carbonized singly, yet their blends with 20 percent of low-volatile Pocahontas No. 3 coal yielded cokes with similar physical properties. For example, when carbonized singly, their respective coke-stability indexes ranged from 9 to 31, whereas the indexes of their blends with 20 percent of Pocahontas coal ranged from 44 to 49. Blending with larger proportions of Pocahontas No. 3 coal resulted in only slight additional increase in coke strength. These results show that the strength of coke made from blends cannot be predicted accurately from the cokemaking property of the component coals.

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[60] Reese, Seth T., and Kearney, Naomi W., Injury Experience in the Coking Industry, 1952: Bureau of Mines Bull. 548, 1955, 20 pp.

[61] Reese, Seth T., and Kearney, Naomi W., Injury Experience in the Coking Industry. 1953: Bureau of Mines Bull. 553, 1955, 18 pp.

Bell County. - Dean-bed coal contained a moderately high percentage of sulfur and yielded weak coke. Mason and Lower Hance coals were chemically suitable for metallurgical purposes; their cokes were similar to those obtained from coking coals of equal rank.

Tennessee Coals

Campbell County. - Red Ash-bed coal was chemically satisfactory for metallurgical use, but its coke was very weak.

Reports on the carbonizing properties of Tennessee coals from Overton, Sequatchie, and Anderson Counties were published. Two high-volatile A Wilder-bed coals from Overton County are too high in sulfur for metallurgical use as mined,[62] but if amenable to preparation, both could be used as minor components of coking blends. Sewanee-bed coal from Sequatchie County ranks high in the high-volatile group and qualifies for metallurgical use because it yields strong coke of moderate sulfur content.[63] Anderson County coals yield highly fissured coke,[64] except that from the Coal Creek bed, which is low in both sulfur and ash and strongly coking. Pewee, Poplar Creek, and Windrock coals probably would be suitable for metallurgical use if blended with larger proportions of higher ranking coals. Big Mary coal is high in both ash and sulfur and yields weaker coke than other coals from this county.

Pennsylvania Coals

Westmoreland County. - Pittsburgh, Redstone, and Upper Freeport coals all had moderately high sulfur contents, which is characteristic of most of the remaining reserves of this county. Pittsburgh coal ranked highest and yielded the strongest coke.

Fayette County. - Sewickley-bed coal was too high in both ash and sulfur to meet present standards for coking coals. Samples of Lower Kittanning from two mines were of medium-volatile rank and qualify chemically for metallurgical use; their cokes had excellent physical properties.

Jefferson County. - Upper Freeport and Lower Kittanning are high-volatile A bituminous coals. Although Upper Freeport yielded strong coke, it probably would be used only as a minor constituent of coking blends because it is high in sulfur. Lower Kittanning yielded coke with relatively low hardness.

Cambria County. - Medium-volatile Upper Freeport and Lower Freeport coals yielded strong coke, in accord with their rank. Both are quite suitable for metallurgical use. Upper Freeport and Lower Freeport from two other mines are low-volatile coals. Although the former is moderately high in both ash and sulfur, it is

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[62] See work cited in footnote 12.
[63] See work cited in footnote 13.
[64] See work cited in footnote 16.

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suitable for blending with larger proportions of lower ranking coals, as shown by tests of its blend with 80 percent of Pittsburgh coal. The latter coal was low in ash and sulfur and yielded coke with excellent physical properties.

Formosa

A blend of 20 percent of Mo Shan with equal portions of Shih Ti, Shin Po, and Sui San coals yielded very weak, fingery coke that was weaker than coke made from 100 percent of Lower Sunnyside, Utah, coal.

Tests With Tuscaloosa Oven

Since establishment of the carbonization section at the Southern Experiment Station, Tuscaloosa, Ala., in 1946, research has been directed along two principal lines: First, a study of coke in its own right and divorced from both coke oven and blast furnace; and second, a study of the effects of operating variables on the carbonizing process and on the quality of coke produced. A third line of investigation, which is expected to be considered in future, is a study of the relation between coke quality and blast-furnace operation. The results of these investigations should contribute to the technology of the iron and steel industry that is vital to the Nation's defense. Long-term benefits include preservation of coal reserves by establishing optimum conditions of operation for coke-oven and blast-furnace installations, thereby bringing about more efficient utilization of raw materials.

The early phases of the investigation were concerned principally with studies of the precision of standard methods for determining the physical properties of coke, together with attempts to devise new testing methods or modify existing methods to obtain more precise data. Results of these studies suggested that physical tests involving a screen-analysis determination as the final step have inherent limitations as to reproducibility. To clarify this picture, an investigation of size-distribution patterns and breakage processes was undertaken.[65] Although several interesting findings with reference to the mechanism of various breakage processes were noted, the most significant conclusion was probably that little is to be gained by minor modifications of existing test methods for determining the physical properties of coke.

More recent efforts have been directed largely toward pilot-scale studies with the Tuscaloosa oven to determine the effects of operating variables (for example, flue temperature, bulk density, type of coal or blend used, and type and amount of inert substance in blend) on the pressure developed during carbonization and the quality of the resultant cokes. Following adoption of a set of standardized operating and testing procedures, an investigation of the reproducibility of data thus obtained was undertaken to provide the necessary statistical parameters needed for evaluating results of future investigations of the effects of operating variables on coke quality.[66] By an analysis of the data for 17 replicate carbonizing tests on a single base blend, precision indexes were established for samples from single and successive lots of experimental coke. By comparing these indexes with those determined for commercial cokes and with those computed from theoretical considerations, it was concluded that, within the limits of sensitivity of the methods of investigation used: (1) Coke from a single experimental-oven test is essentially uniform with respect to physical properties, and (2) the uniformity of cokes from different sources probably decreases in the following order: A single experimental charge, a single commercial charge, successive experimental charges, and successive commercial charges. In addition to the data on reproducibility, the report includes results of tests of several coals carbonized at different temperatures and a summary of recent test data for coals and blends carbonized, using established operating and testing procedures.

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[65] Gayle, J. B., and Auvil, H. S., Studies of Size-Distribution Patterns and Breakage Processes for Metallurgical Cokes: Bureau of Mines Rept. of Investigations 5096, 1954, 40 pp.

[66] Gayle, J. B., and Eddy, W. H., Carbonizing Tests With Tuscaloosa Oven, 1952-53: Bureau of Mines Rept. of Investigations 5125, 1955, 16 pp.

Because most coals do not exert measurable pressures on the walls of the Tuscaloosa oven when carbonized under conditions of moisture and bulk density approximating those used by commercial plants in this district and because the practice of drying samples before testing creates operational difficulties at this station and, since it may influence the quality of coke produced, two sole-heated ovens were constructed as supplementary equipment for studies of coal expansion. To date, these ovens have been used for testing duplicate samples of coals carbonized in the Tuscaloosa oven, for surveying the expansion characteristics of selected coals and coal blends, and for studying factors influencing coal expansion. In addition, the results of physical tests on sole-oven cokes have been used to obtain information as to factors influencing the quality of coke in slot-type ovens.

Effect of Anthrafines on Expansion of Coking Coals

The results of an investigation of the effects of incorporating varying amounts of anthrafines into a given base coal or blend are of particular interest.[67] As a preliminary part of this investigation, the merits of the sole-heated oven as a tool for studying coal expansion were considered briefly. Using a new method of interpretation, it was shown that results for slot-type and sole-heated ovens are influenced similarly by variations in charge density, probably the most important variable influencing the expansion of a given coal. This finding is particularly encouraging in that it suggests that other similarities between results obtained with slot-type and sole-heated ovens may be found.

Having concluded that sole-oven results are more directly comparable with those for slot-type ovens than normally considered, tests were conducted on blends containing varying proportions of anthrafines and other inerts. Results of these tests indicate that the particular inert substances studied are highly effective in reducing the expanding tendencies of coking coals of widely varying rank; and, using the new method of interpreting sole-oven data, it was estimated that additions of anthrafines amounting to 5 percent result in decreases of some 30 to 50 percent in wall pressures.

Although it was pointed out that the results suggest that the effectiveness of any solid inert in reducing coal expansion is proportional to its density, this point could not be definitely established; in fact, the data available indicated that expansion for a given coal is determined principally by the inert-free, dry-coal charge density. Continuing work on this problem (not published) has fully confirmed the first of these statements and clarified the situation with reference to the second. Thus, although the density of the particular inert used is of considerable importance, most of the tests reported were for blends containing anthrafines and coke breeze that probably do not differ greatly in this respect when crushed to the sizes used for blending. More recent studies have shown that blending moderate amounts of anthrafines and similar materials (apparent specific gravity, about 1.7) decreases expansion, whereas certain types of chars (apparent specific gravity, about 0.9) have little effect, and other chars and certain insulating materials (apparent specific gravity, about 0.3) actually show increased expansion when the results are compared at the same charge density.

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[67] Gayle, J. B., and Eddy, W. H., Effect of Anthrafines on Expansion of Coking Coals: Bureau of Mines Rept. of Investigations 5176, 1955, 18 pp.

Low-Temperature Carbonization of Noncoking Coals

Studies on the drying and low-temperature carbonization of the noncoking coals have been under way for several years at the Bureau's Denver laboratory. Several industries, particularly the electric-power industry, can operate more efficiently if supplied an upgraded fuel from low-rank western coals. It has been demonstrated that power can be generaged more efficiently from either dried coal or low-temperature char than from raw, high-moisture coal. Tar obtained as a byproduct from low-temperature carbonization of the noncoking coals is potentially a valuable material in the chemical industry, since it contains a mixture of hydrocarbons and oxygenated compounds used in the solvent and plastics fields. Upgrading low-rank coals may make them more attractive as fuel for thermal power generation in competition with oil and natural gas.

Complete facilities for pilot-plant testing of the drying and carbonization properties of noncoking coals are available. These units are designed to process the coal in the entrained and fluidized state and have capacities up to 1 ton per hour. Valuable experience in the design and operation of a large industrial plant has been obtained through observing a 240,000-kw. powerplant utilizing the drying scheme developed at Denver.

Application of this type of processing on an industrial scale was described in a recent publication.[68] It was demonstrated that the economics of applying low-temperature carbonization on a large scale depends to considerable extent upon the yield and potential value of tar. The coal when carbonized produces tar, gas, and char. Most of the gas will be used to supply the heat of processing, and the char will be used as fuel for the steam-generating plant. The tar therefore must pay for the processing operation plus the value of the potential heat in the raw coal that has been converted to and appears as tar and gas. Depending upon the rank of coal, the potential heat in the char will range from 75 to 80 percent of the potential heat of the raw coal, and the tar must pay for the remaining 20 to 25 percent. The margin that this will leave for cost of processing, as related to the cost of delivered coal, is shown in figure 13. Texas lignite upon carbonization yields 17 gallons of tar per ton, while some Utah coal produces 44 gallons. Because of the higher tar yield of Utah coal, it is possible to spend more on its processing. For example, if a Texas lignite costs $2.00 per ton delivered at the plant and yields tar having a value of 10 cents a gallon, $1.25 could be spent for processing. On the other hand, Utah coal costing $5.00 per ton would yield enough tar selling for 10 cents a gallon to permit expenditure of $3.00 per ton in processing costs.

Carbonization Studies of Western Coking Coals

A study in cooperation with the western steel industry was instituted during the latter part of 1955 to study the carbonizing properties of Colorado and Utah coals available for producing metallurgical coke and developing means for improving the quality of such coke. These coals are in rather limited reserve and in addition

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[68] Parry, V. F., Low-Temperature Carbonization of Coal and Lignite for Industrial Uses: Bureau of Mines Rept. of Investigations 5123, 1955, 27 pp.

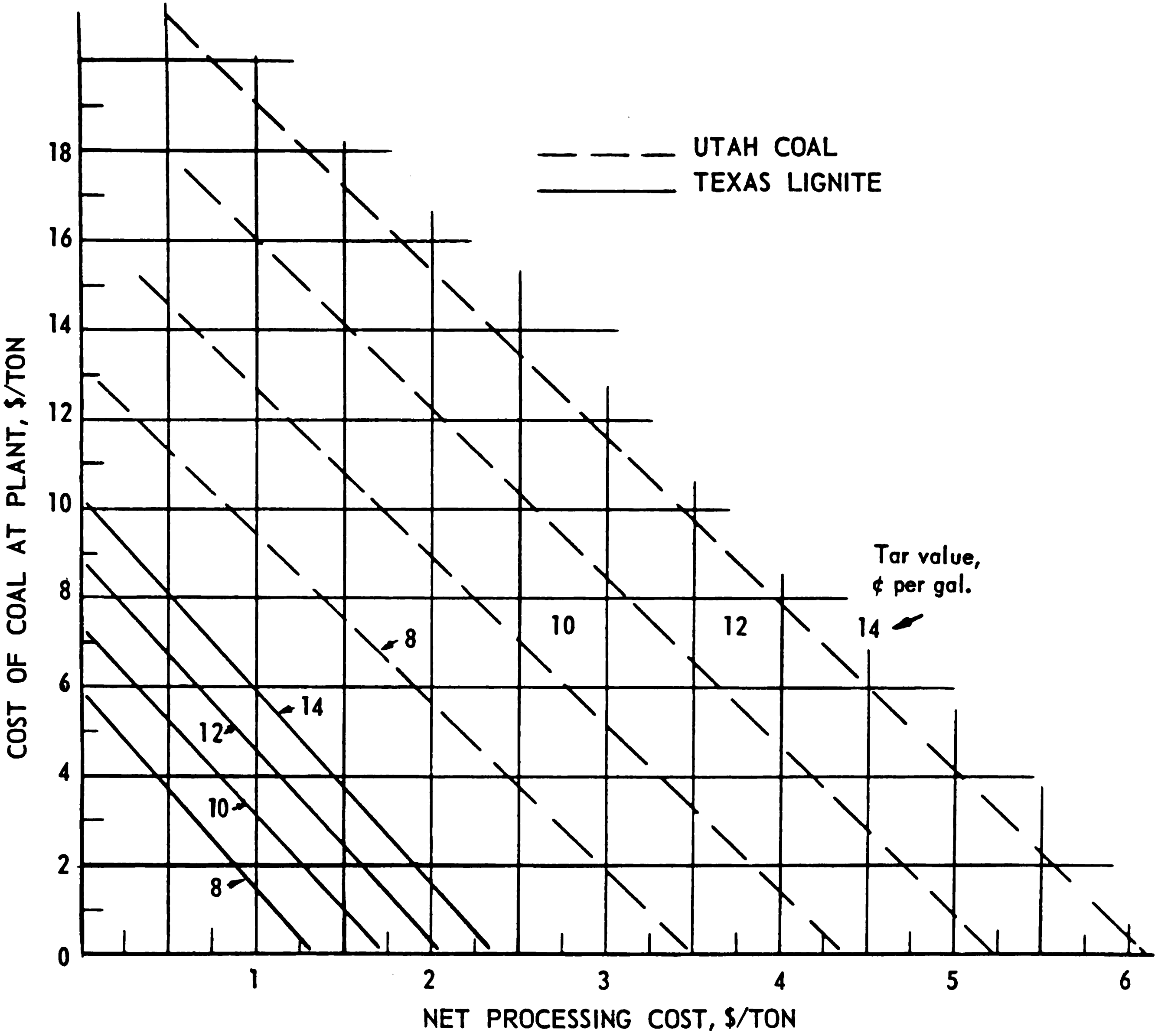

Figure 13. - Cost relationship when coal is processed for powerplants.

have poor coking qualities. The western steel industry has the problem of increasing the potential supplies of coking coals, the production of improved coke by use of blends with higher quality coals, and obtaining improved quality and uniformity of coke through preparation. Each of the three major western steel producers has been actively conducting a research program on this problem; but it has been recognized that a coordinated research study, with particular emphasis on the application of petrography for determining the composition of coking coal and as an aid in developing preparation flowsheets, would be valuable. The studies outlined will consist of establishing the carbonizing properties of the major coking coals of the western area by several laboratory tests, to correlate such properties with the petrographic composition of the coals, and to apply such petrographic information to preparation problems.

Four samples representing coals from Colorado and Utah are being carefully protected in the laboratory to prevent oxidation and loss of coking properties. Representative samples of each coal are being carbonized under various conditions to determine the effect of bulk density, size, and admixtures on the quality of the coke. Petrographic analyses will be made by both reflected- and transmitted-light techniques, and an attempt will be made to develop a rapid scheme for preparing representative samples of whole coal for such analyses. Small-scale laboratory tests are also used to establish the caking and plastic properties of the coals, giving a rapid means of evaluating changes produced through preparation. The preparation studies in connection with this problem are being conducted in cooperation with the Research Foundation of the Colorado School of Mines. It is anticipated that completion of this study will require several years.

Low-Temperature-Tar Research

In recent years interest in low-temperature carbonization has increased owing to successful application of the fluidized-bed techniques that had been proved so economically advantageous in the petroleum industry. Fluidized carbonization may offer a new market for coal (especially lignite and other low-grade coals) in connection with an integrated electric-power and tar industry, in which the char from the carbonization would be used as boiler fuel and the tar would be the raw material for a new chemical industry. The establishment, on a large scale, of an integrated electric power-tar industry depends on determining tar components, their possible utilization, and their upgrading to more economically attractive materials. Research on low-temperature tar therefore has three main objectives: (1) Separation and characterization of fractions and components from commercial and experimental low-temperature tars; (2) economic upgrading of these tars, and various fractions and components, to produce chemicals of greater value; and (3) evaluation of all of these various fractions and components for utilization as plastics, resins, fuels, lubricants, paints, pesticides, etc.

A low-temperature-tar project was started at the Appalachian Experiment Station, Morgantown, W. Va., at midyear, and the laboratory is being equipped for it. Meanwhile, the tars derived from lignite are being studied at the Lignite Experiment Station, Grand Forks, N. Dak., and tars from both lignite and subbituminous coals are under investigation at the Denver Station.

At the Lignite Experiment Station the work thus far is a qualitative examination of one commercial tar to establish procedures for investigating tars obtained from other sources and/or under different conditions of carbonization and recovery. The tar was supplied in the form of a distillate and pitch fraction. The distillate has been separated further by conventional acid-alkali extraction into tar acids, tar bases, and a neutral oil. Other methods of gross separation are being investigated, including the Duo-sol process of liquid-liquid countercurrent extraction. The tar acids are separated by distillation into comparatively simpler fractions that are examined by chemical and physical methods.

Interest is centered currently in the tar-acid fraction that comprises 51.5 percent of the distillate (tar bases 1.25 percent, neutral oil 47.25 percent) from this particular tar. The largest part of this material is high-boiling (above 210° C.), and characterization is made rather difficult by the lack of background information in this range. Phenol, cresols, and xylenols have been isolated in appreciable quantity; pyrocatechol, resorcinol, guaiacol, and naphthols have also been separated.

Preliminary investigation of the neutral oil by silica-gel separation indicates that it is also largely high-boiling material, predominantly aromatic, and contains appreciable quantities of neutral heterocyclics. This fraction is insoluble in 20-percent acid or alkali and distills to 300° C. without appreciable decomposition.

Infrared-absorption methods have provided a powerful tool to supplement chemical methods in the tar-evaluation program. During the year a single-beam double-pass instrument was obtained, and personnel experienced in research applications of infrared absorption techniques was recruited. Examination of some narrow-boiling fractions has established the presence of m- and p-cresol, 2,3-xylenol, 2,4-xylenol, 2,5-xylenol, m- and p-ethyl phenol, and guaiacol. An important phase of this program is compilation of a catalog to include spectra of all possible phenolic homologs; to accomplish this will require additional work in synthesis and purification where compounds are not available or are of poor quality. This is necessary to permit identification as well as quantitative estimation of the phenolic materials present, since comparable curves are not available for the compounds suspected to be present; enough information is now available for making reasonable quantitative estimates regarding some of the lower boiling tar acids in the distillate fractions.

Fractional crystallization of liquid tar-acid fractions at greatly reduced temperatures effected the separation of a crystalline compound which, on examination of its spectrum in the 3-micron region, appears to be an alkyl-substituted dihydroxy aromatic for which no comparable spectra were found. This example accentuates the importance of this program to collect spectra of known compounds. A number of phenolics are on hand, and the work is concentrating on those suspected to be present in the tar-acid fraction currently under investigation.

At the Denver Station the effects of heat, oxidation, and storage on the properties of tar are being studied. Normally these agents have little influence on high-temperature byproduct tars, since such tars are chemically more stable. On the other hand, it has been demonstrated that low-temperature tar contains more complex and chemically active compounds that are affected by the dual phenomena of oxidation and autopolymerization. Previous work has shown also that the properties of primary low-temperature tars are altered with time. This effect is believed to occur in some instances without the sustaining external influence of air, light, or heat.

Present work shows that aging and oxidative processes produce pitch at the expense of the normally distillable fraction. That fraction of the distillate boiling about 270° C. is mainly involved in pitch formation due to storage, while all fractions are affected to some degree when the tar is heated at 125° C. in the presence of air. If the effects of air are considered, the aromatic hydrocarbons and tar-acid fractions appear to be involved in the pitch formation.

The experimental results indicate further that pitch can form through the normal aging process as a result of polymerization reactions among the unsaturated-hydrocarbon compounds, with some contribution from the tar acids. The first effect would appear to reduce the neutral oil, while the second seems to reduce the total tar acids. Experimentally, these considerations have been found to hold true for short periods of oxidation and storage.

The American literature has been singularly deficient in presenting data on the properties of primary low-temperature tars. A review of the pertinent foreign and American work has been made. This information, combined with the original laboratory data on low-temperature tars collected by Bureau workers, constitutes the basis for publications on low-temperature tar that are being issued. In one of these the general knowledge on low-temperature tar is summarized. It traces the character of coal tar from incipient formation, through the condensation of the tar vapors, to the subsequent alteration of tar products by further heating. Data are presented to support the view that the tar yield bears a definite relation to coal rank, which, in turn, is a function of the oxygen content of the coal substance.

The literature study indicates that tar exists as a complex mixture of hydrocarbon material whose general composition is governed by the thermal process used to liberate it from the coal, and the constitution of the parent coal. Present knowledge of low-temperature and high-temperature tars from byproduct processes indicates that the incipient tar fragments, as liberated, probably were large and that these have undergone both degradation and synthesis on exposure to heat. This could account for the appearance of the complex, highly substituted tar constituents at the lower temperatures of carbonization. Characteristically, it has been shown that the greater proportion of the complex, individual entities constituting the tar mass appears only in trace amounts.

Twenty typical low-temperature tars obtained from carbonization of various ranks of domestic and foreign coals in the fluidized pilot-plant retort have been analyzed and reported in the other publication. The data indicate that, in carbonization techniques employing entrainment or fluidization of the coal particle, the carbonization conditions are secondary to coal rank with respect to tar yields and composition. There is wide variation in the composition of low-temperature tars obtained by the carbonization of low-rank coals. The composition of such tars obtained in the pilot plant from coals ranging in rank from lignite through high-volatile C bituminous are shown in table 2.

Effects of Process Variables in Coal Carbonization

Data obtained in the Survey of Carbonizing Properties of American Coals, which was initiated 26 years ago in cooperation with the American Gas Association, were studied to show the dependency of carbonization on process variables. The results of this study, which have been published,[69] show numerous similarities among bituminous coals of various ranks with respect to their behavior when these variables are changed. Effects of coking temperature, coal-bulk density, and blending on the yield of products, strength of coke, and expansion can be predicted from a single carbonizing test on a high-volatile coal. Variations in temperature and bulk density exert considerable influence on the uniformity of coke. For variations of a given magnitude (about 200° C. in temperature and about 4 pounds per cubic foot in bulk density) the hardness of coke will vary about 15 percent. This variation in the quality of coke exists for all ranks of Appalachian coals.

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[69] Smith, F. W., and Reynolds, D. A., Effect of Temperature, Charge Density, and Blending on the Carbonization of Coal: Bureau of Mines Rept. of Investigations 5110, 1955, 24 pp.

TABLE 2. - Average properties of low-temperature tar derived from fluidized carbonization of low-rank coals at 800 to 1,000° F.

Specific gravity, dry tar	60°/60°	0.96 - 1.15
Solidifying point	°C.	25° - 30°
Distillation to 300° C.	percent dry tar	30 - 65
Pitch, distillation to 300° C.	do.	35 - 70
Composition of distillate, volume-percent of distillate to 300° C.:		
0° - 170°C.		5.0
170 - 200°C.		8.0
200 - 210°C.		5.0
210 - 235°C.		15.0
235 - 270°C.		20.0
270 - 300°C.		40.0
Pitch residue and loss		7.0
Constituents of distillate, volume-percent:		
Acids		20 - 65
Bases		1 - 5
Neutral oil		30 - 75
Olefins		30 - 70
Aromatics		15 - 40
Paraffins		15 - 35

Effects of Preheating on Carbonizing Properties of Coal

The effects of preheating on the carbonizing properties of coal are being studied with the objective of developing a method of improving the cokemaking property of high-oxygen coals, thereby rendering them more suitable for manufacturing metallurgical coke. Most midwestern and western coals yield weaker coke than Appalachian coals; hence their value for metallurgical use would be greatly enhanced by a suitable beneficiating process. Furthermore, the process probably could be used to restore, at least in part, the coking properties of eastern coals depreciated through weathering.

Preheating at temperatures below the fusion point has been found to increase the strength of the coke, particularly if there is no intermediate cooling before carbonization. Preheating alters the basic chemical structure of coal and affects the yields of carbonization products under some conditions. A report describing this work was prepared and presented at the 1955 annual meeting of the American Institute of Mining and Metallurgical Engineers.

Coal Expansion During Carbonization

The Bureau of Mines sole-heated oven is used to determine the expanding properties of coals and blends. Only coals with more than 65 percent dry, mineral-matter-free fixed carbon are tested, because most coals of lower rank contract during carbonization. These tests are of particular interest to coke-oven operators because knowledge of the expansion of coals and blends is necessary in commercial coking to prevent oven damage.

Pratt-, Mary Lee-, and Pittsburgh-bed coals were tested because pushing difficulties were encountered occasionally when a blend of these coals was carbonized commercially. The expansion of Pratt coal, which was found to have increased since it was tested several years ago, probably accounts for the pushing difficulties.

A blend of four Formosan coals contracted in the sole-heated oven and therefore should not expand during carbonization in commercial ovens.

Bench-Scale Tester

Development work on a bench-scale expansion tester, using only 71 grams of coal, was continued. The ultimate objective is to develop a test method using a small quantity of coal and requiring less time for testing than the sole-heated oven. Tests are being made to establish a correlation with the sole-heated expansion oven that used 40 pounds of coal per test. If satisfactory correlation is accomplished, the smaller unit will supplant the sole-heated oven for routine testing, thereby effecting considerable savings in equipment and operating cost. The standard deviation of the test precision for this apparatus, determined from 29 tests of high-, medium-, and low-volatile coals, was ± 1.7 percentage points.

The sensitivity of the bench-scale tester to changes in blending ratios was determined by testing Pittsburgh and Pocahontas No. 3 coals and their blends. The linear relationship between the proportion of low-volatile coal used and expansion was found to be fair; a 10-percent change in the proportion of low-volatile coal altered the expansion about 3 percent. Reproducibility in duplicate bench-scale tests of single coal types, splint, semisplint, and bright coal, was no better than is obtained on coals sampled as usual.

A correlation study between the bench-scale tester and the sole-heated oven is in progress. Fifteen coals have been tested; 2 to 5 bench-scale tests were made on each coal, but only 1 sole-oven test was made because the variability of the latter has been determined. A comparison of the average results obtained by the two methods shows that the bench-scale tester exaggerates the expansion of coals as their expansion in the sole-heated oven increases.

Mechanism and Kinetics of Coal Carbonization

Studies of the mechanism and kinetics of coal carbonization, initiated last year to furnish fundamental data on coal carbonization and to develop new techniques for assessing the factors controlling coking, were continued. A study of the thermal decomposition of high-molecular-weight materials was initiated as another project in this general investigation.

Effect of Coal Fluidity on Coking

One of several projects continued in studying the mechanism and kinetics of coking involves the fluidity of coals and their blends in the plastic state attained in carbonization. Initial studies were made, using high-melting-point pitches with Gieseler maximum fluidities up to 60,000 dial divisions per minute, to provide a wider range in fluidity than is attainable with coal alone. The fluidity of each pitch and that of a Pittsburgh-bed coal was reduced progressively by blending with finely pulverized coke. Carbonization was carried out under a pressure of 2 pounds per square inch. The abradability of the resulting solid residues (chars) was unusual, because, as the proportion of coal was increased to 35 percent, the abradability index or hardness increased progressively to a maximum, then decreased to a minimum at 65 percent coal, and again increased to a maximum at 95 to 100 percent coal. These unusual changes in the abradability of char occurred when the same blends were carbonized at higher pressure, although the maximum and minimum indexes differed from those of chars made at the normal pressure. Other tests, wherein various sizes of pulverized coke were blended, showed that only the finest size (minus-325-mesh) could be used in large proportions without lowering the abradability index markedly.

At this stage of the investigation, certain sizes of inerts also appear to be more effective than others in stabilizing foam produced during the fluid state of carbonization. Probably there is an optimum coke size, intermediate between the finest and largest used in these tests, that increases the stability of foam most effectively. The larger sizes contribute to foam stability and the chars from their blends exhibit the characteristic porosity at an intermediate coal-coke ratio. Apparently, the finest size is too small to effectively stabilize the foam system; therefore, a more compact and less abradable char is formed.

Thermal Decomposition of High-Molecular-Weight Materials

The composition of coal tar is related to the composition of the coal when carbonization is effected at low temperatures; but, under high-temperature carbonizing conditions, the influence of coal composition is largely overcome by pyrolysis of the compounds, which explains the characteristic differences in low-temperature tar. The study of the formation of tar and its alteration in escaping from the coke oven has such fundamental importance that a thermal cracking unit was constructed to evaluate the effects of the variables involved. Tests in this apparatus should provide basic knowledge on the coking process with which the formation of tar is intimately associated.

The fluidized-bed tar generator using nitrogen as the fluidizing medium provides tar vapors for analysis and further thermal processing at approximately the same temperature as that at which tar is formed in commercial coke ovens. Tests are thus made available for systematic study at temperatures ranging from that of their formation to those encountered in passing from the plastic layer through the hotter coke and contacting the hot oven walls. These tar vapors are then subjected to controlled conditions of temperature, residence time, ambient atmosphere, and surface area in a thermal cracker to simulate conditions in oven coking. Examination of the decomposition products and tests of the effect of temperature on tar composition promise to increase knowledge of the mechanism and kinetics of tar formation at various coking temperatures. Furthermore, the data obtained may be of value in determining the path of travel of gases and vapors in the coke oven.

Other variables can be determined with little or no change in design. The tar generator, as well as the thermal cracker, can be operated to determine the effects of temperature, atmosphere, pressure, and catalyst. The heat for carbonization, which is derived in part from the fluidizing gas, is supplemented by heating the carbonizer or tar generator externally.

Analyses of tars produced without the cracking stage, during about 10 to 15 minutes residence time in the generator, show that the maximum temperature to which they were exposed is equivalent to 500° to 600° C. in the BM-AGA retort. Heat-transfer rates to the char-coal-carbonizing mixture through the wall were approximately 50 B.t.u. per hour, per square foot, per degree Fahrenheit.

Vapor-Phase Reactions of Thermal Decomposition Products of Coal

Vapor-phase reactions of sulfur with the liquid products from the thermal decomposition of coal are being studied as a method of determining the mechanism and kinetics of coking and the formation of liquid and gaseous products. Investigation of vapor-phase reactions of sulfur with benzene show that they go to completion to form carbon disulfide and hydrogen sulfide, although the yields obtained have not exceeded 27 percent of theoretical owing to a loss in catalyst activity. Intermediate products are obtainable by reducing the reaction time.

These data will be correlated with those from reactions of sulfur with hydrocarbons of the benzenoid and naphthalene series, and also polynuclear hydrocarbons.

Kinetic data from these studies will aid in controlling the variables when coal tar fractions are used as the hydrocarbon in the reaction. Carbon disulfide or intermediate-reaction products probably could be produced from the less valuable coal-tar fractions by this method.

Pretreatment of Coal

Investigation of the effects of pretreatment on the carbonizing behavior of coal promises to yield worthwhile information on the mechanism of coking. The testing of Pittsburgh coal was continued, with emphasis on the effect of controlled oxidation at elevated temperatures on its coking property. This coal, 1/8 by 3/8 inch in size, was oxidized continuously without ignition, using a shallow bed and a superficial air velocity of 9 feet per second at 250°, 300°, 350°, 375°, and 390° C. until its coke-making property, as measured by small-scale abrasion and shatter tests, was virtually destroyed.

The reduction in coking property of Pittsburgh coal by air oxidation is a function of the oxidation time and temperature. The strength of coke, as evaluated by the abrasion test, is constant or is increased somewhat at the start of oxidation and then decreases sharply with additional oxidation. The fluidity of the coal is decreased below the minimum required to make strong coke by extensive oxidation. As expected, the coking property of this Pittsburgh coal persists longest at the lowest oxidation temperatures and decreases very rapidly at higher temperatures; it is destroyed after 5 hours of oxidation at 375° C. or 3 hours at 390° C., as compared with 50 hours at 250° C. The sharpest decline in the coking property occurs between 350° and 390° C., which is within the range of active decomposition of this coal under these experimental conditions. At these temperatures thermal decomposition, rather than oxidation, probably is primarily responsible for the rapid reduction in coal fluidity to the minimum required to form strong coke. The decrease in coking power depends less upon increased oxygen content per se than on changes in the constitution of the coal.

The volatile-matter content of Pittsburgh coal decreases initially upon oxidation at all temperatures used in this investigation; it increases on further oxidation at 200° to 350° C. The decrease is most marked in the 350° to 390° C. range because this coal is actively decomposed at these higher temperatures. The increase in volatile matter at the lower oxidizing temperatures is accounted for by addition of oxygen to form a coal-oxygen complex, which decomposes into carbon dioxide and water during the volatile-matter test.

Oxidized 1/8- by 3/8-inch coal was agglomerated slightly during carbonization by exuded fluid material. This fluid material was shown by microscopic examination to be present to some extent, even after prolonged oxidation, at all temperatures. However, the "inert" surfaces of the oxidized coal particles are not wetted by this material; therefore, cohesion is weak, and the strength of the coke decreases rapidly with oxidation. This effect is in contrast with the results obtained when the oxidized coal is crushed before carbonization, in which instance the fresh surfaces exposed are wetted by the exuded material still present in enough quantity to agglomerate the particles and make a strong coke.

The hydrogen content of coal and its density, or specific volume, are linearly related. This linear relationship is not changed significantly by oxidation, even though the hydrogen content is lowered. Such a relationship indicates a single type of molecular structure and packing common to oxidized and unoxidized coals.

Carbonization of Lignite in Hydrogen Atmosphere

As a preliminary step to obtain design information for a fixed-bed gasification unit to be built at the Lignite Experiment Station, lignite was carbonized at 935° F. and pressures of 600 to 1,000 p.s.i.g. in a hydrogen atmosphere. The results provide information on the degree of agglomeration that may be encountered in the gas generator.

Although distinct agglomeration was obtained at 1,000 p.s.i.g., as shown in figure 14, it was not observed at pressures below 600 p.s.i.g. and therefore is not expected to cause difficulty in the gas generator. In the hydrogen atmosphere, recovery of solid residue decreased with increasing pressure, ranging down to 50 percent of the moisture- and ash-free lignite at 1,000 p.s.i.g., due to hydrogenation.

COAL AND LIGNITE GASIFICATION

Conversion of Coal to High-B.t.u. Gas

Because of the increased demands for natural gas, both for industrial and domestic consumption, and the fact that this natural resource is not an inexhaustible commodity, it has become increasingly apparent that the possibility of obtaining fuel gas from alternative sources should be considered. With this in view, the Bureau has actively engaged in a study of making high-B.t.u. gas from coal. The coal is first gasified to synthesis gas, (a mixture of carbon monoxide and hydrogen), and this gas is then catalytically converted to methane. The methanation of synthesis gas in fixed-bed, conventional fluidized-bed, and multiple-feed, fluidized-bed equipment has been studied to develop suitable catalysts and processes.[70/] The results show: (a) Methanation reactions may be carried out in 1-inch, bench-scale equipment at high synthesis-gas throughputs with good temperature control of the reactions if a multiple-feed type fluidized-bed design is used; (b) nickel is superior to iron as a catalyst for the methanation of synthesis gas, unless the process is considered in terms of a combined operation that produces both gaseous and liquid products; (c) techniques of catalyst preparation determine, to a large extent, the process of life of nickel catalysts, and these techniques must be investigated thoroughly before any conclusions can be reached; and (d) enough data are available to indicate that the concentrations of sulfur in synthesis gas used for methanation are extremely critical, and maximum process limits should be evaluated carefully.

Recycling Residue in Entrainment-Type Gasifier

The results of investigations by the Bureau and others during the past few years have demonstrated that it is feasible to produce $CO + H_2$ by continuous gasification of pulverized coal with oxygen and steam. However, it has been found that, if enough oxygen is used to gasify the coal completely, the excessive oxygen requirement per unit volume of synthesis gas produced makes the process uneconomical. As a possible method for reducing the cost of synthesis gas and completely gasifying the carbon in the coal, the effect of recycling the unburned residue in a vortex gasifier was investigated. The results of this investigation indicated that

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[70/] Greyson, Murray, Demeter, J. J., Schlesinger, M. D., Johnson, G. E., Jonakin, James, and Myers, J. W., Synthesis of Methane: Bureau of Mines Rept. of Investigations 5137, 1955, 50 pp.

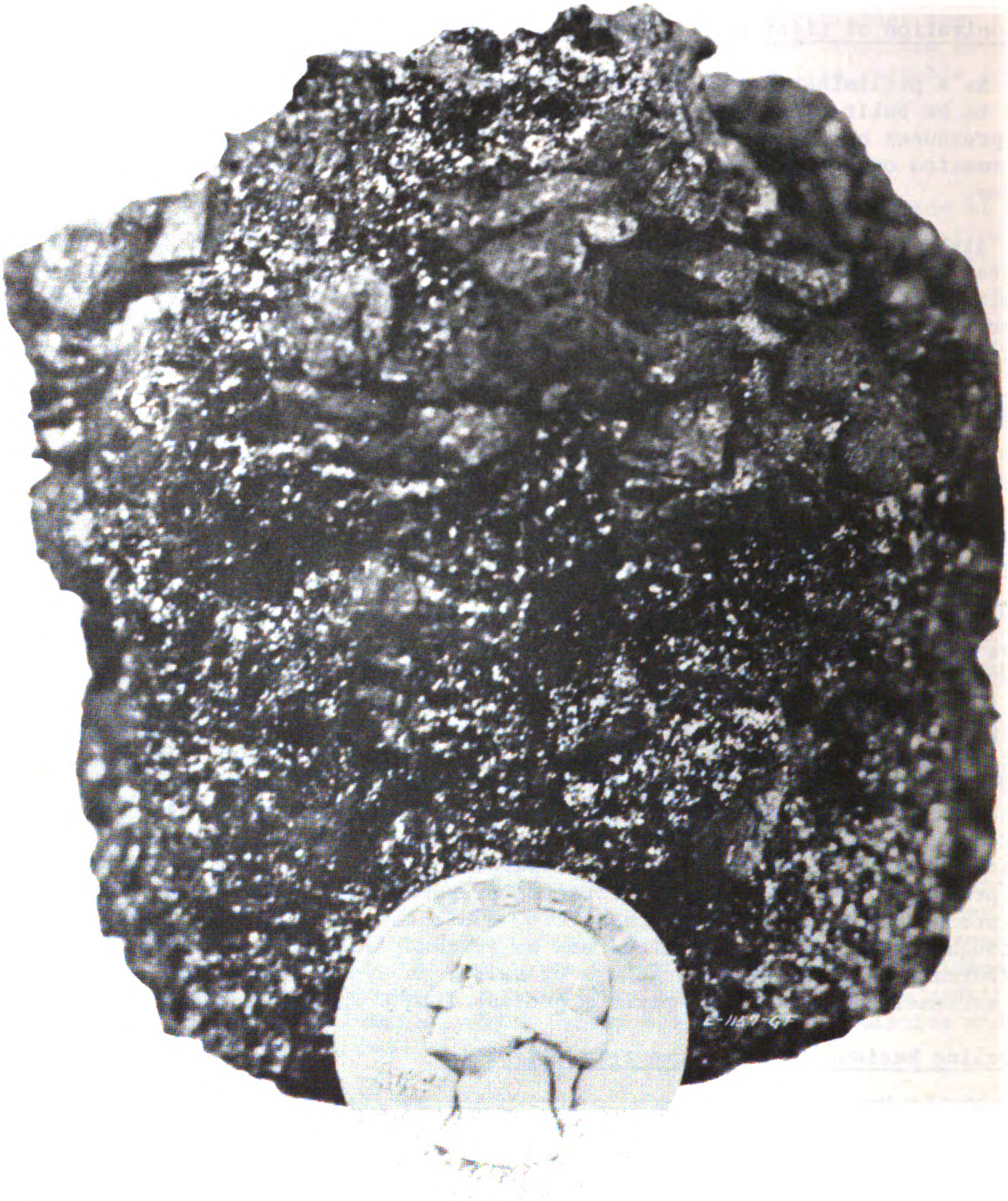

Figure 14. - Solid residue from carbonization of North Dakota lignite in hydrogen atmosphere at 1,000 p.s.i.g. and 935° F. Diameter of coin, 1 inch.

recycling the unburned residue is a promising method for increasing the efficiency of the entrainment-type gasification process. In addition, if used in conjunction with slagging operation, the ash-disposal problem is reduced to a minimum.

Study of Various Gasification Processes

A paper study is being conducted of various methods of generating a high-B.t.u. gas that will be interchangeable with pipeline gas. All of these studies are based on the use of coal as the basic raw material for high-B.t.u. gas production. The size of the plant under study was set at 90,000,000 std. cu. ft. per day, which represents approximately 0.1 percent of the total quantity of natural gas sold today.

Preliminary flowsheets were prepared for the following proposed processes:

1. Nuclear gasification of coal at 325 p.s.i.g., plus methanation of synthesis gas at 300 p.s.i.g. to produce a 916-B.t.u. gas.

2. Oxygen gasification of coal at 325 p.s.i.g., plus methanation of synthesis gas at 300 p.s.i.g. to produce a 925-B.t.u. gas.

3. Nuclear gasification of char at 325 p.s.i.g., plus hydrogenation of coal at 300 p.s.i.g. to produce a 972-B.t.u. gas.

4. Nuclear gasification of char at 450 p.s.i.g., plus hydrogenation of coal at 1,200 p.s.i.g. to produce a 965-B.t.u. gas.

5. Oxygen gasification of char at 325 p.s.i.g., plus hydrogenation of coal at 300 p.s.i.g. to produce a 951-B.t.u. gas.

Raw-material requirements, which include nuclear fuel requirements where applicable, were determined from flowsheets. These studies indicate that gasification plus hydrogenation may be the most economical method of producing high-B.t.u. gas from coal.

Pilot-Plant Gasification of Lignite

The fifth and final report in a series covering earlier pilot-plant gasification of lignite in the annular-retort gasifier at Grand Forks, N. Dak., has been published.[71] The pilot plant was operated over 8,700 hours, with the primary objective of obtaining information on the mechanical performance of the heat-transfer surface of the externally heated retort. Data on the effect of process variables and design modifications also were obtained.

Underground Gasification

Field-scale, underground gasification experiments at Gorgas, Ala., are continuing in an effort to develop means for recovering low-cost heating or synthesis gas from coal beds that are uneconomical to mine.

Underground gasification requires the production of passages through the coal to introduce air and recover gas. Early experiments with developing such passages by mining and by "electrolinking" have been followed by a method of hydraulic fracturing in which the coal is fractured by hydraulic pressure and the passages so created are held open by sand grains carried in the fracturing liquid. An 80-fold increase in the permeability of the beds has been obtained with this method, and fracturing around a borehole extends for a radius of 200 to 300 feet.

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[71] Oppelt, W. H., Kube, W. R., Chetrick, M. H., Kamps, T. W., and Golob, E. F., Gasification of Lignite in a Commercial-Scale Pilot Plant, Progress Report from July 1, 1950 to December 31, 1951 and Summary of Work Previous to July 1, 1950: Bureau of Mines Rept. of Investigations 5164, 1955, 80 pp.

Two methods of gasification were tried during the year. In the first, designated as forward or down stream, air was introduced at the ignition point and passed through the fire area, and the gases were conducted through the fractured coal to the point of recovery. This method of operation proved unsuccessful. Deposition of tar in the fractures and expansion of the coal beyond the fire area as it was heated by the products of combustion produced high resistance to gas flow. The rate of gas output amounted to less than 10 percent of the air-input rate.

In the second method, designated as backward or upstream, the direction of air flow was reversed; air was introduced through the fractured coal and passed through to the fire area, and the products of combustion were evacuated through the coked or burned-out region of the bed. Tar deposition and expansion of the unburned coal were eliminated, thus maintaining the permeability of the system. Gas recovery represented 65 percent of the air input. The average analysis of the gas produced was as follows:

	Analysis, percent
Carbon dioxide	10.7
Oxygen	.3
Illuminants	.4
Hydrogen	8.0
Carbon monoxide	11.8
Methane	2.8
Nitrogen	66.0
Heating value, B.t.u. per cu. ft. 60 F., 30 in. Hg, dry	100
Specific gravity	0.95

Test conditions did not permit the high air-injection rate believed necessary for complete gasification. As a result, only 13 percent of the coal affected was completely gasified, while the other 87 percent was just carbonized; however, the results obtained are encouraging, and further tests under more favorable conditions are under way.

RESEARCH ON PRODUCTION AND PURIFICATION OF SYNTHESIS GAS FROM COAL

Pilot-Plant and Bench-Scale Studies

Coal Gasification

The aim of this program is to develop low-cost processes for gasifying coal. Gas mixtures thus made may consist mainly of carbon monoxide and hydrogen for the synthesis of ammonia, liquid fuels, and other organic compounds; or they may contain these gases and various gaseous hydrocarbons and thus be suitable substitutes for natural gas. The development of gasification processes involves also evolution of suitable methods and equipment for feeding the reactants - coal, steam, and oxygen - to the gasification equipment.

In July 1954 the laboratories were moved to the new Appalachian Experiment Station in West Virginia. The remaining months of 1954 and most of 1955 were spent in rebuilding the pilot plants and installing auxiliary equipment. A part of the facilities for pilot-plant work at the new station is shown in figure 15. Research was not resumed until late in 1955, and no new results can be reported now. However, data obtained in the atmospheric-pressure and high-pressure pilot plants during 1953-54 have been evaluated, and detailed publications are in process. The tests in these two plants were made according to a factorial design to permit a statistical analysis of the results. The relative significance of various factors was established, so that the less important parameters may be eliminated in future work.

Subbituminous C coal, high-volatile bituminous A coal, and anthracite were gasified with oxygen and steam in the atmospheric-pressure gasifier at various coal-feed rates and oxygen:carbon and steam:carbon ratios. The extent of carbon gasification was highest for the subbituminous C (Lake DeSmet) coal and lowest for the anthracite with the same carbon input rate and reactant ratios, as shown in figure 16. This effect of rank on gasification is ascribed to differences in (a) flame geometry, which will vary with the volatile content of the fuel; (b) the physical and chemical properties of the fuel particles, as related to their reactivity with steam and CO_2; and (c) the mean effective temperature in the reactor, which is determined by these factors and by heat losses from the reactor.

In both this high-pressure and atmospheric-pressure work it was established that the oxygen requirement increases about 3.0 cu. ft. for each 1,000-B.t.u. increase in heat loss.

Operation of the high-pressure, water-cooled gasifier was directed toward determining the effect of residence time on the extent of gasification and on the oxygen and coal requirements per unit volume of $CO + H_2$ at various pressures and throughputs (pounds of coal per hour per cubic foot of reaction volume). The average loss of heat in this gasifier was approximately 36 B.t.u. per std. cu. ft. of $CO + H_2$ produced. Sewickley coal (bituminous A) was used at 4 pressures, 3 feed rates, 3 oxygen:coal ratios, and 1 steam:coal ratio. Residence time affected the extent of gasification.

The high-pressure gasifier is now lined with silicon carbide, so that less heat will be lost than in the previous design. Data obtained in the old and new reaction chambers will be combined to relate the effects of heat loss and residence time to gasification.

Coal-Feeding Methods

Feeding methods must be devised for transferring fine coal into high-pressure gasifiers. Up to now a pneumatic feeder developed by the Bureau has been used. In this feeder fine coal is fluidized with a gas, and its flow rate is measured by the pressure drop across the feedline. At high pressures it is expensive to compress fluidizing gas.

Currently, 2 other methods of feeding fine coal at high pressures (about 30 atmospheres) are being investigated in pilot plants designed to feed up to 1,500 pounds of coal per hour. In one of these, finely divided coal is mixed with water and then pumped through a coil heater. The resulting steam-coal mixture can be introduced into the gasifier at about 1,000° F. Pumping costs are comparatively low, and good control of the feed rate is possible; however, the slurry contains more

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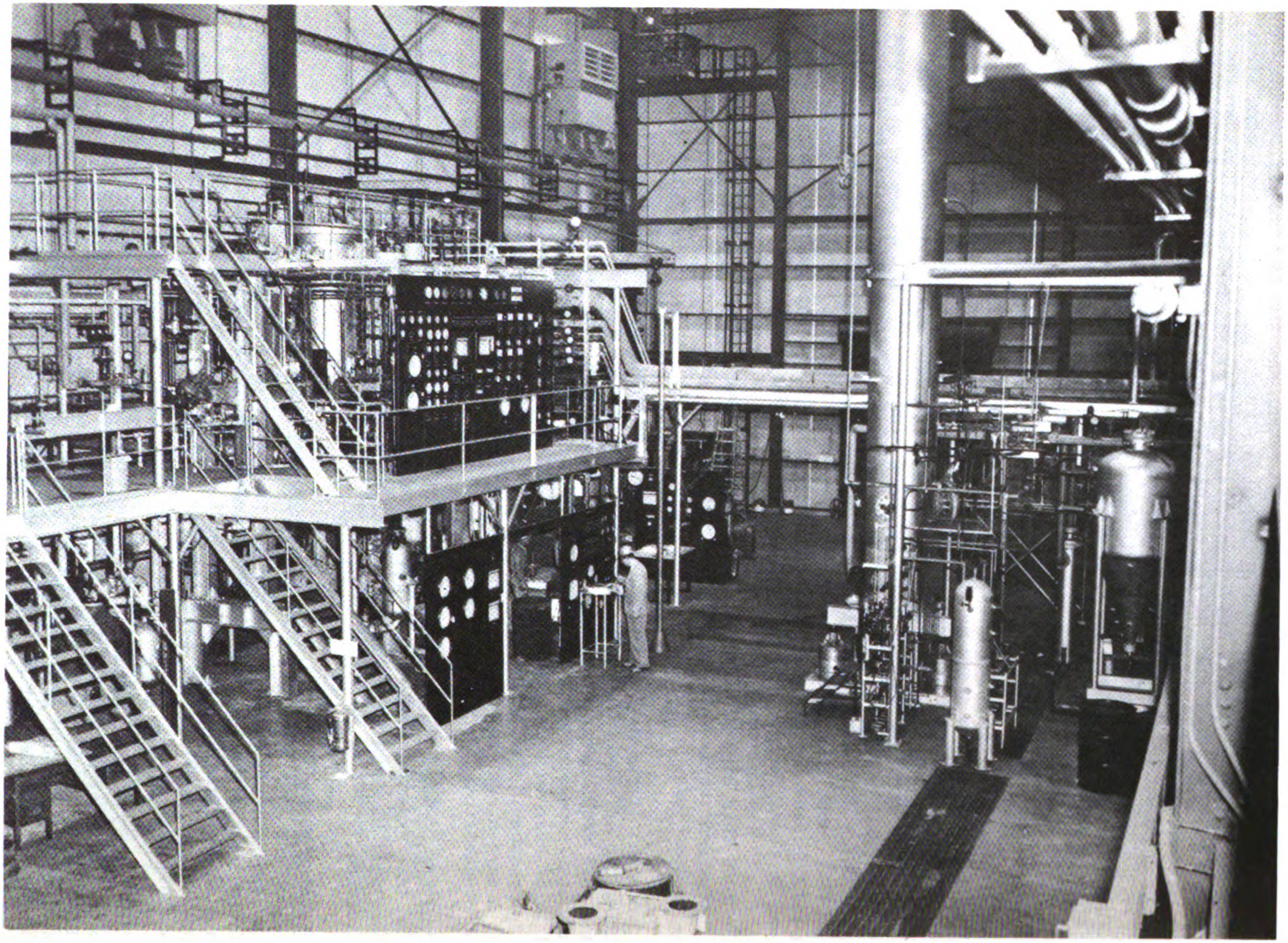

Figure 15. - Part of facilities at Bureau of Mines Appalachian Experiment Station, Morgantown, W. Va. for pilot-plant work. Pressure gasifier No. 3 in left foreground.

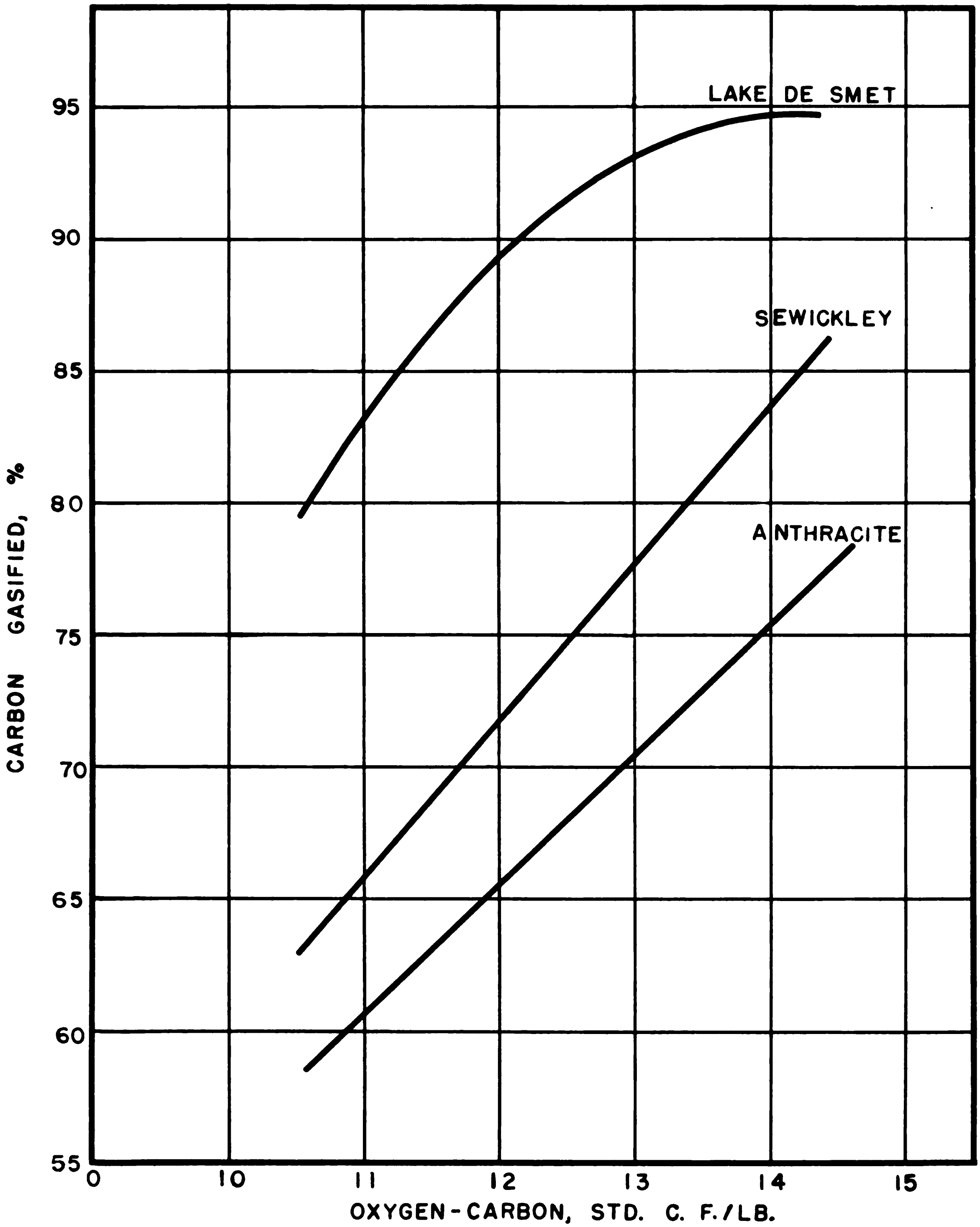

Figure 16. - Comparison of percentage of carbon gasified for Lake DeSmet and Sewickley-bed coals and anthracite at a constant steam-carbon ratio of 0.70 lb. per lb. and at a constant carbon input of 430 lb. per hr.; atmospheric gasifier.

steam than is desirable. A steam-coal separator must therefore be developed to reduce the proportion of steam (to about 0.5 to 1 part per part of coal) and to recover the steam in usable form. Current work is concerned with heater designs to minimize erosion of coils. The work to date has centered on this problem and also on development of methods of determining the flow rate by measurement of the viscosity or density of the coal-water mixture.

In the other method, an attempt is being made to introduce finely divided coal under pressure at a measured rate into a high-pressure, superheated steamline. Such a method has been used at low pressures; at high pressures, however, several problems need solution if the method is to be economical. A screw feeder is being developed for use at 30 to 40 atmospheres. Concurrent problems are adaptation of feeders to take coal from the pressure vessel and development of a suitable injector for the steam pickup line. The carrying capacity (that is, ratio of coal to steam) of superheated steam and reheating of the steam-coal mixture must be investigated.

Gas Purification

Coal gasification produces a raw synthesis gas that must be purified to meet specifications established for various syntheses. One method of producing gas, which meets tentative specifications established for the Fischer-Tropsch synthesis, consists of four steps: (1) Removing dust by scrubbing with water, followed by electrostatic precipitation; (2) removing carbon dioxide and most of the hydrogen sulfide by scrubbing with an aqueous solution of diethanolamine; (3) removing hydrogen sulfide by iron oxide; and (4) adsorbing organic sulfur compounds by activated carbon. Investigations are being made to improve this process and develop other processes that will lower the purification cost.

Ion-exchange resins are being studied as a means for removing gaseous impurities (COS, H_2S, and CO_2). Experiments were made at atmospheric pressure and 300 p.s.i.g. with Amberlite IRA-400 resin to remove carbonyl sulfide in a 48-inch ion-exchange column of 1/2-inch, type 304, stainless-steel, Schedule 40 pipe.

In 1 experiment 0.36 cu. ft. per hour of mixture of carrier gas and 12.4 grains of sulfur as COS per 100 std. cu. ft. were passed through the column (bed height, 34 inches) at 300 p.s.i.g. Effluent gas was analyzed periodically, so that the breakthrough point (0.1 gr. per 100 std. cu. ft.) could be ascertained. Under these conditions about 1.7 std. cu. ft. of gas was purified per gram of ion-exchange resin. Under a cooperative agreement between the Bureau and The General American Transportation Corp., small-scale pilot-plant experiments are being made to ascertain the possibility of reducing the cost of removing carbon dioxide and hydrogen sulfide by replacing the conventional packed column with a turbomixer-type gas-liquid contactor. This agitated liquid-gas contactor consists of a 42-inch length of 18-inch, Schedule 80, carbon-steel pipe, 9-inch-I.D. lift tube, with 4 vertical baffles 1 inch wide and equally spaced, lift impeller, annular ring, and saturator impeller (see fig. 17). A 1-hp. motor (1,750-r.p.m.) is used to drive the impeller at shaft speeds of 500 to 900 r.p.m.

Experiments have been made with a 40-percent solution of diethanolamine and inert gas containing 10 to 12 percent of CO_2 per gallon at a pressure of 250 p.s.i.g. The shaft speed for these experiments was maintained at about 550 r.p.m. More CO_2 was absorbed in this type of absorber than either a packed or bubble-cap column. Runs at 760 and 970 r.p.m. caused vortexing and bypassing of feed into the foul-solution outlet line. Relocation of the outlet to the side of the vessel eliminated this condition. Experiments with a 60-percent solution of diethanolamine at the

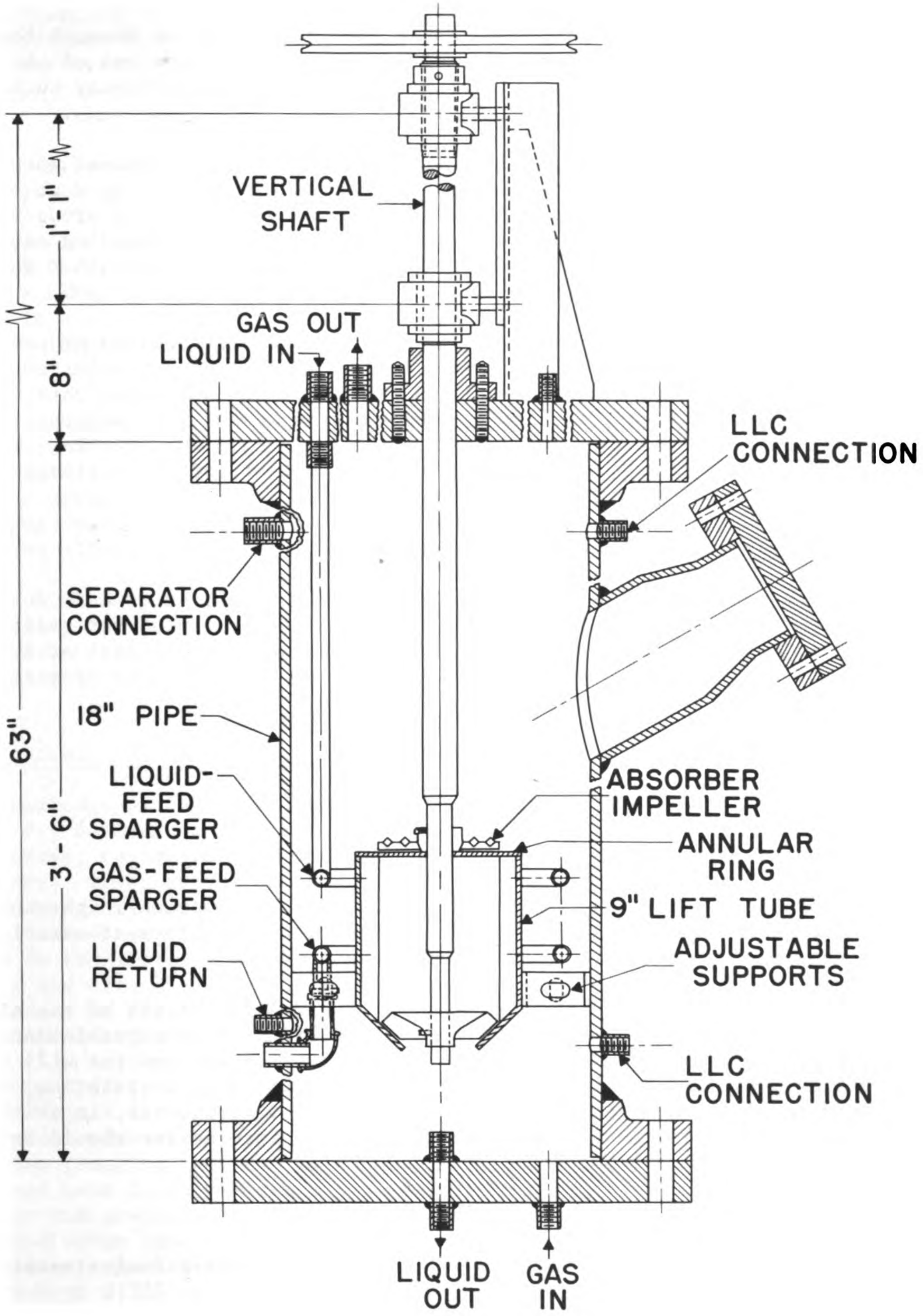

Figure 17. - Turbo-contactor for gas absorption.

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same pressure were run after a sightglass was installed to permit visual observation of flow patterns at various speeds. Some of the feed gas passed up through the lift tube, a condition that should be prevented. To direct the feed gas out of the throat of the impeller and distribute it uniformly under the liquid spray curtain, the gas inlet was redesigned as shown in figure 17.

Entrained dust must also be removed from raw synthesis gas. Present work on this problem involves studying dust sizing and preparing and developing dust-removal equipment. A counterflow feeder now being developed may be useful for producing low concentrations of very fine particles. The effectiveness of dust-removing equipment in the pilot plant is studied whenever possible. The packed water scrubber was particularly effective at high pressure.

For proper evaluation of dust-removing equipment, the distribution of particle sizes in the gas stream should be known, since the type of equipment selected will depend in large measure on such size distribution. A method for making this determination is nearing completion. The gas is passed through a plastic membrane filter that retains all particles and agglomerates at the point where they strike. Variations of this method may also prove satisfactory for determining the ultimate particle size, or even dust concentration.

Use of Nuclear Energy to Supply Heat for Coal Gasification

Only recently have nuclear reactions been considered a source of heat for high-temperature chemical reactions. A system in which the heat required for gasifying coal and steam is supplied by nuclear energy has the following potential advantages over one in which partial combustion of coal with oxygen furnishes the necessary heat:

1. Elimination of oxygen plant and compressors.

2. Reduction of coal requirements by 40 percent.

3. Improvement of gas quality due to lower CO_2 and H_2S content.

A preliminary cost estimate showed that a substantial reduction in the cost of producing synthesis gas could be attained if the formidable problems of materials, design, construction, operation, and maintenance can be solved.

Solution of these problems is being undertaken as a joint effort of the Atomic Energy Commission and the Bureau of Mines. The Bureau is testing suitable materials for constructing the reactor-fuel elements. The interior of the reactor will be subjected to slag abrasion; and the reactants will be exposed to irradiation, since they may flow directly through the reactor. From a simulated reactor, in which electric heat is substituted for nuclear heat, the following information should be obtained:

1. Choice of suitable materials for construction.

2. Effect of slag erosion on materials proposed for nuclear fuel-element cladding.

3. Effect of geometric design of fuel elements on flow of slag.

4. Optimum temperature of reactor, a compromise between higher temperatures that speed gasification and aid the flow of slag, and lower temperatures that prolong the life of refractory materials.

5. Optimum steam-coal ratio for best gas yield.

6. Rates of heat transfer that may be tolerated with various cladding materials.

One simulated reactor design is shown in figure 18. This design consists of a 4-inch-I.D. silicon carbide tube, packed with 1-1/2-inch silicon carbide-coated graphite spheres. A coil carrying a high-frequency current surrounds the reaction tube. To reduce the heat radiation to the high-frequency coil, one-half inch of granular insulation is provided between the reaction tube and the coil.

The coil consists of 1/2- by 3/4-inch rectangular copper tubing and is water-cooled. The high-frequency input will be from a 100-kw., 9,600-c.p.s. motor generator.

The design rate at atmospheric pressure is 10 pounds of coal and 15 pounds of steam per hour. This gives an approximate linear velocity of 8 feet per second, making no allowance for slag buildup in the passage between spheres.

It will be possible to test single balls up to 3-1/2 inches in diameter. One of the major objectives will be to ascertain the heat flux possible with various sizes of spheres clad with different coatings.

Laboratory-Scale Research

Kinetic Study of Steam-Carbon Reaction

Knowledge of the mechanism and rates of reaction at high temperatures (up to 3,500° F.) between superheated steam and carbon of various types and grade (graphites, cokes, coals) would be helpful in developing and designing large-scale coal gasifiers. Temperatures above 3,000° F. have been recorded in the gasifiers operated in Morgantown, and there is reason to believe (from failure of the gasifier linings and other refractory materials) that much higher flame temperatures are obtained in the vicinity of the burners. Such information would also help in interpreting and extending the present knowledge about the steam-carbon reaction. To obtain data on the reaction mechanism and rates, a high-temperature isothermal gasification reactor was built for kinetic studies up to 3,500° F.

During design and construction of the high-temperature reactor an earlier reactor designed for operation up to 2,400° F. had been used to determine the effect of particle size on the gasification of electrode graphite by steam. Closely sized particles (ranging from 60- to 230-mesh) were dropped at a constant rate of 4 to 5 grams per hour from a vibrated feeder bowl through a refractory feed tube of 5-mm. diameter and preheated to the reactor temperature by the time they emerged. Excess preheated steam (about 10 cu. ft. per hour) entered through the top and contacted the narrow stream of carbon particles at the end of the carbon feed tube. Carbon and steam at 2,400° F. continued in parallel downflow toward the reactor exit, where the residue was collected in a glass receiver. Residual carbon was weighed and again passed through the reactor. Each run consisted of 4 or 5 passes.

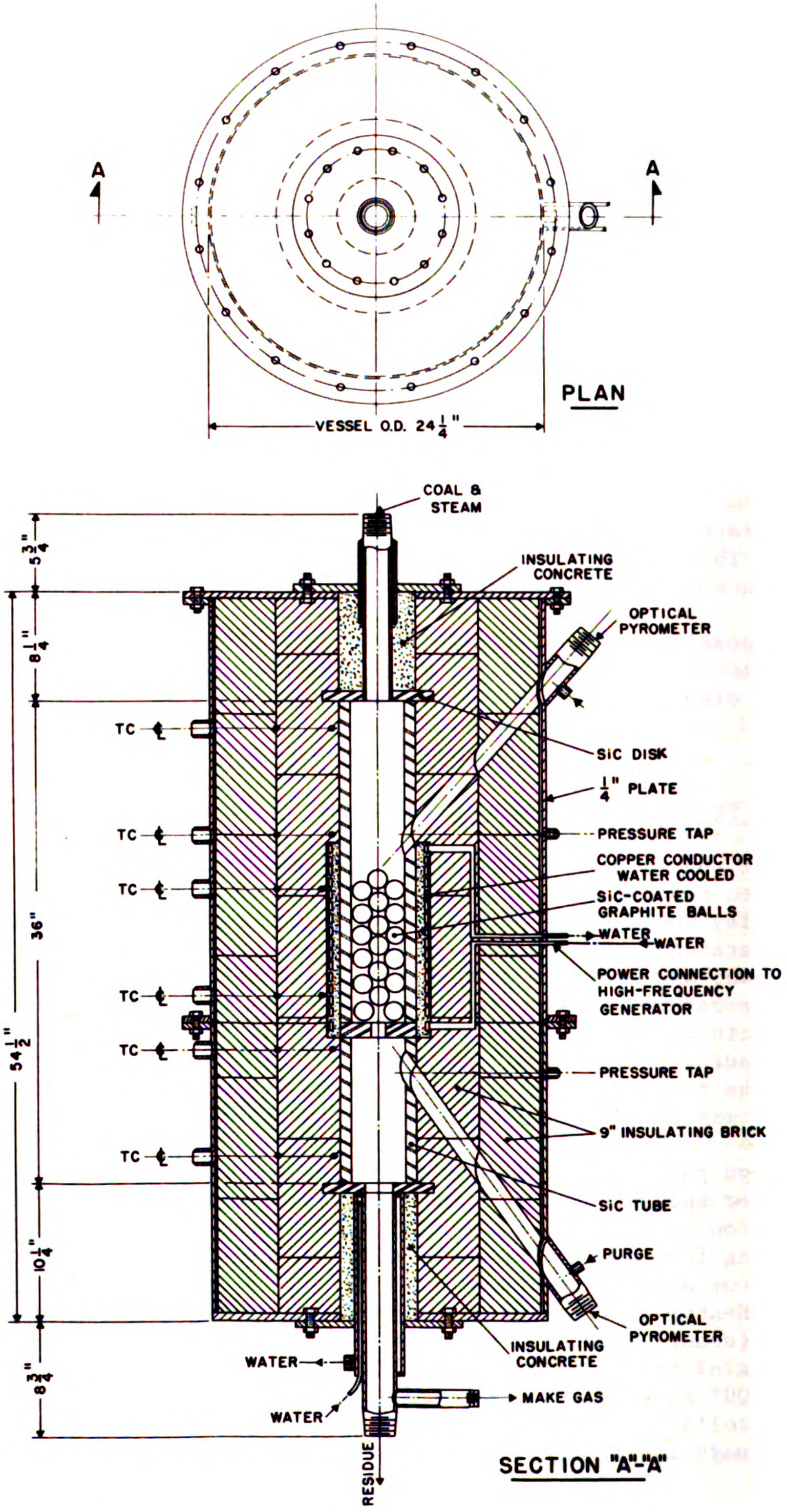

Figure 18. - Simulated nuclear reactor IBR (1).

Tests made in this 3-inch-I.D. by 6-1/2-foot-long, electrically heated, vertical, tubular reactor, previously described,[72] showed that the rate of reaction with steam was a linear function of particle diameter. A decrease of 100 microns in the latter caused about a 6-percent increase in carbon conversion. The relatively high initial rate decreased rapidly with increasing contact time up to 10 seconds. Further evaluation of the data is in progress. Microscopic and X-ray studies of the original and partly converted particles are being made.

Flashing of Coal-Water Slurries

The purpose of this work has been to develop a method of rapidly vaporizing (flashing) coal-water slurries in an externally heated alloy tube coil into a suspension of fine coal in steam. Information from this study should be useful for designing a coal feeder that may have advantages over the pneumatic methods now used for feeding powdered coal.

Approximately 25 runs have been made with an anthracite and subbituminous coal in a bench-scale apparatus for flash-vaporizing coal slurries. Pressures up to 150 p.s.i.g. and temperatures up to 750° F. had no effect other than to change the velocity of steam through the 50-foot-long, 3/8-inch-diameter coil. The results indicate that flashing itself caused no breakup of particles. Micronization was due to impact and abrasion, which increase with large increase in velocity, changes in flow direction, and centrifugal force. Considerable increase in velocity, the chief cause of micronization, has been achieved by decreasing the pressure at the coil outlet.

Gasification of Coal Slurries in a Heated, Metallic, Tube-Coil Reactor

The purpose of this study was to obtain information needed for developing a process for gasifying coal with steam but without oxygen. This method is especially desirable when waste heat at 1,600° to 1,800° F. is available (for example, in graphite piles of atomic-energy installations), although heat from nuclear fission reactions at temperatures up to 4,000° F. might also be used if available. The experimental procedure consists in pumping the coal-water slurry at a constant hourly rate of 1 to 5 pounds of coal into a 117-foot-long, 3/8-inch, alloy-steel tube coil of 9-inch coil diameter. Current is passed directly through the coil to heat it. The reaction products and excess steam are passed through a condenser and scrubbing vessels; the gases are then passed through a sampling and metering system and vented.

An early run had indicated that 66-percent carbon conversion can be obtained at about 1,800° F., despite a very short contact time, but the alloy tube had a short life. At 1,850° F. outlet-gas temperature, approximately 83 percent of the carbon in Lake DeSmet, Wyo., coal was gasified in a contact time of one-half second. The gas produced contained about 16 percent of carbon monoxide, 60 percent of hydrogen, 20 percent of carbon dioxide, and 3.5 percent of methane. Carbon conversion was a linear function of both residence time and gasification temperature.

Problems Related to Use of Nuclear Energy for Producing Synthesis Gas

This investigation parallels the study of gasification of coal-water slurries by nuclear energy. Laboratory-scale research was begun on the corrosion and erosion

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[72] Bureau of Mines, Synthetic Liquid Fuels, Annual Report of the Secretary of the Interior for 1954; part I - Oil From Coal: Rept. of Investigations 5118, 1955, p. 50.

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resistance of materials of construction and on determining methods and mediums of heat transfer. These and other problems must be solved before such a process can become practical.

A small, simulated, tubular reactor was constructed. The 26-inch-long, 2-inch-I.D., silicon carbide reactor tube was electrically heated by a carbon-spiral resistor up to 3,000° F. It was filled with 3/4-inch-diameter silicon carbide balls to reproduce, on a small scale, the conditions that would be encountered in nucleonic reactors where heat for the steam-carbon reaction is available from radioactive material enclosed in silicon carbide balls.

Powdered coal was charged into a large mixer tank in which the slurry was prepared. The coal-water slurry was pumped (at a rate of 1 to 3 pounds of coal per hour) through a tube-coil furnace in which it was preheated to 1,000° to 1,500° F. The resulting coal-steam suspension was passed downward through the reactor; and molten slag was to be tapped at the bottom. Gas and excess steam were passed through two condensers and scrubbing vessels; the gases were then passed through a sampling and metering system and vented. One run was completed with a subbituminous coal.

The apparatus may also yield information on reaction rates and reactant ratios. Plans have been made to study the properties of various materials, such as silicon carbide, zirconia, and zirconium carbide, in a solar furnace to determine their suitability for use in high-temperature gasification reactors.

Metal Chelates as Oxygen Carriers

The objective of this investigation is to develop an economical process for producing tonnage oxygen from air, based on selective sorption of oxygen by a chemical oxygen-carrier, and subsequent recovery by desorption. The basic problem is to find a stable compound with suitable sorption properties.

To be competitive with the currently used rectifying of liquefied air for producing oxygen, a new process must satisfy the following conditions: (1) Relatively small temperature difference between the absorption and desorption steps; (2) low heats of reaction for both oxygenation and reduction steps; (3) absorption of oxygen from the air at pressures below 75 p.s.i.a. and desorption at pressures above 1.0 inch of Hg; (4) simple apparatus; (5) virtually no deterioration of the sorbent on cycling.

Salcomine has been shown to meet all these conditions except slow deterioration on cycling. Although it deteriorates only 0.025 percent per cycle, this loss of activity is enough to make the process uneconomical. Thus, the problem is to prevent or reduce this deterioration significantly or to develop a more stable compound. Deterioration may be attributable to the loss, during cycling, of a water molecule present as a bridging group between two Salcomine molecules.

Studies are planned on the nature and mechanism of oxygen sorption by Salcomine and its several derivatives; on removal of peroxide, ozone, and other impurities from air; and on elimination of impurities occluded in Salcomine crystals, which may cause irreversible "oxidation" instead of the desired reversible "oxygenation."

SYNTHESIS OF LIQUID FUELS FROM HYDROGEN AND CARBON MONOXIDE (FISCHER-TROPSCH AND RELATED PROCESSES)

Pilot-Plant and Bench-Scale Research

Hot-Gas-Recycle Process

Previous process development on the Fischer-Tropsch reaction by the Bureau of Mines was restricted to liquid-phase systems. The oil-circulation or slurry processes are suitable for producing a range of hydrocarbon products at lower synthesis temperatures; however, when high yields of gasoline are desired, temperatures of around 300° C. are preferable, for which liquid-phase processes are not well suited. Therefore, an investigation was begun of the hot-gas-recycle system in which the heat of reaction is removed by circulating gas rather than by oil.

A deterrent to previous development of this process was the high pressure drop of gas through a bed of granular catalyst. The power expended in circulating large volumes of gas would be prohibitive unless the catalyst beds were rather shallow. Development of steel lathe turnings has now provided a catalyst with physical and chemical stability that allows a relatively low pressure drop because of the high void space. As a result, the cost of circulating gas through a 10-foot bed (22 cents per gallon of product for fused granular iron (6- to 20-mesh) can be lowered to 0.5 cent with turnings having 92 percent of void space.

A pilot plant for investigating the hot-gas-recycle process has been constructed and is now in operation with lathe turnings prepared from S.A.E. 1018 carbon steel (see fig. 19). The 6-foot bed contains 92 percent void space and permits a pressure drop of only 8.5 p.s.i., in agreement with previous bench-scale studies. Synthesis was started with $1.3H_2+1CO$ gas, 400 p.s.i.g., and a 60:1 recycle ratio. From 70 to 90 percent of the gas has been converted at 305° C., with an hourly space velocity (fresh gas) of 1,000. The maximum space velocity obtained in the oil-circulation reactor with the same catalyst was 600.

Synthesis With Nitrided-Iron Catalysts in Oil-Circulation and Fluidized Reactors

The application of nitrided-iron catalysts to the Fischer-Tropsch synthesis has already been demonstrated in fixed-bed and slurry reactors. A high yield of oxygenated compounds, especially of alcohols, was produced with nitrided catalysts. In the slurry system, in which the finely pulverized catalyst is suspended in a cooling oil, 21 to 45 percent of the total weight of oil (C_3 + hydrocarbons + oxygenates) was oxygenated organic compounds. The lower molecular weight alcohols can be extracted and later reblended for octane improvement after bauxite treatment of the resulting gasoline fraction. Upgrading techniques have been developed for the various product fractions.[73]

During the past year nitrided-iron catalysts were used in the oil-circulation and dry-bed fluidized systems. In the oil-circulation system the catalyst is immersed in a cooling oil which circulates through the bed to remove the heat evolved in the synthesis reaction. The size of the catalyst particles is much larger than that used in the slurry system. The dry fluidized system employs small particles suspended in a stream of synthesis gas.

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[73] Schlesinger, M. D., and Benson, H. E., Upgrading Fischer-Tropsch Products: Ind. Eng. Chem., vol. 47, October 1955, pp. 2104-2108.

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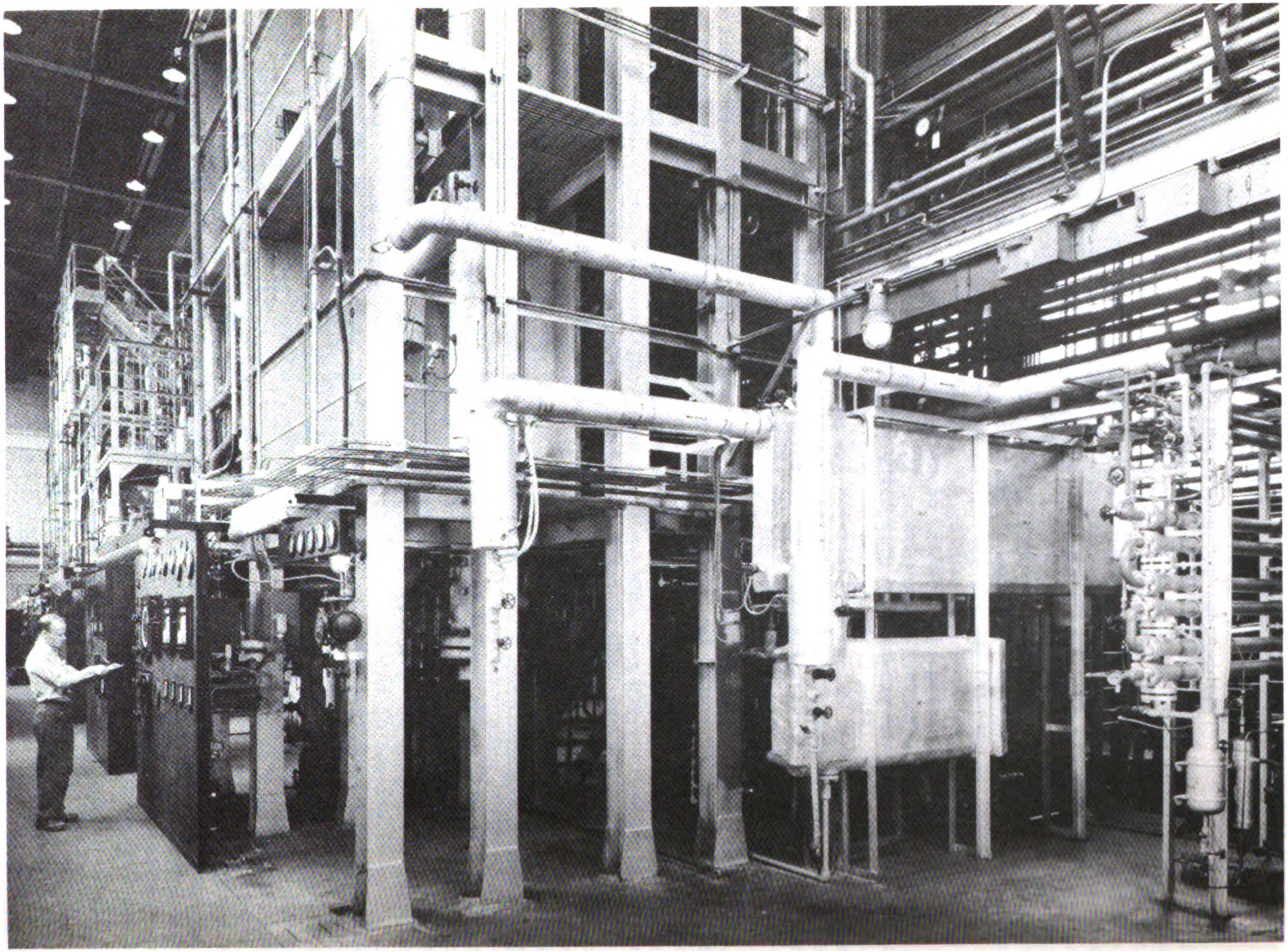

Figure 19. - Hot-gas recycle pilot plant for Fischer-Tropsch synthesis.

The fused nitrided-iron catalyst charged to the oil-circulation reactor was activated at 300 p.s.i.g. and 200° to 220° C. for several hundred hours. The catalyst was extremely stable and was still fairly active when synthesis was voluntarily terminated after 3,700 hours. During that time a series of tests was made to determine the effect of the operating variables upon the yield of oxygenated products. The production of oxygenated compounds was lower than in the slurry system but was improved materially when the oil vaporized from the reactor was not refluxed to the reaction zone. The boiling range of all products was much lower than that produced with the reduced, fused iron oxide catalyst.

The fluidized study was conducted in the same equipment as had been used previously to investigate the synthesis of methane.74/

The fluid-bed reactor consisted of a 1-inch-diameter pipe 6 feet long containing a concentric 3/8-inch baffle tube. Reduction and nitriding were done with the catalyst fluidized. After a short activation period synthesis was initiated at a fresh-feed hourly space velocity of 3,000, with a recycle ratio of 3:1 at 300 p.s.i.g. pressure. During the synthesis several variables were studied. Conversion of synthesis gas averaged about 80 percent, and the condensed liquids, exclusive of water, contained 80 to 90 percent of oxygenated compounds. After 837 hours of continuous operation the experiment was terminated voluntarily. The catalyst had not oxidized, and only a negligible amount of fines was produced. Also, no free carbon was present in the discharged catalyst, even though the partial pressure of hydrogen was only 60 to 90 p.s.i. during the last 500 hours of operation. Comparison of operations with nitrided fused iron in fluidized-bed, slurry, and oil-circulation systems showed that, in pilot-plant equipment, the fluidized bed produced more oxygenated compounds, probably owing to the higher linear velocity and shorter residence time.

Removal of Hydrogen Sulfide by Hot Carbonate Scrubbing

Synthesis gas produced by gasifying coal with steam and oxygen contains an appreciable amount of hydrogen sulfide (0.4 to 0.8 percent), as well as carbon dioxide and small amounts of carbonyl sulfide. Since an extremely low concentration of sulfur compounds poison catalysts for the Fischer-Tropsch synthesis, these impurities must be almost completely removed. Preliminary pilot-plant experiments indicated that simultaneous removal of carbon dioxide and hydrogen sulfide was possible in the hot carbonate system. Small-scale tests have now been made to determine equilibrium pressures of hydrogen sulfide over solutions of potassium carbonate, so that the theoretical efficiency of purification might be calculated. Under conditions similar to those in the pilot plant, equilibrium partial pressures of carbon dioxide were identical to those obtained previously without the presence of hydrogen sulfide. Similarly, water-vapor pressures of the solutions were not affected. At 90° C. the equilibrium pressure of hydrogen sulfide over a regenerated solution (33.3 percent of the potassium carbonate converted to bicarbonate) containing 20 grains of hydrogen sulfide per gallon was 0.38 mm. At a total pressure of 300 p.s.i.g. the equilibrium concentration in the gas phase would be 1.55 grains of hydrogen sulfide per 100 cu. ft. For a more highly regenerated solution the bicarbonate and sulfide contents should be quite low, because the bisulfide is decomposed more readily than the bicarbonate.

74/ Greyson, M., Demeter, J. J., Schlesinger, M. D., Johnson, G. E., Jonakin, J., and Myers, J. W., Synthesis of Methane: Bureau of Mines Rept. of Investigations 5137, 1955, 50 pp.

Studies of Slurry Synthesis Process

A major obstacle to successful operation of many slurry experiments has been settling of the catalyst. Glass reactors, shown in figure 20, were assembled to simulate conditions in the reactor and to observe the action of the catalyst particles. When a 200- to 400-mesh fraction of catalyst particles was suspended in a hydrocarbon oil and heated, a temperature was observed at which the particles dropped rapidly to the bottom of the tube. Lowering the temperature resulted in resuspension of the particles. A few materials have been added in incremental amounts, and the maximum temperature reached before settling was determined. With Bentone-34 a temperature of about 200° C. was reached. A marked difference between fused-iron and iron-ore catalysts was that the former settled at a temperature about 70° higher than the magnetite. This observation suggests that the composition of the surface of the particles is an important factor in maintaining the suspension.

Laboratory Investigations of Gas-Synthesis Process

Development of Catalysts

Studies were continued on the development of fused- and massive-iron catalysts. Twelve difficult reducible oxides were tried as structural promoters in fused catalysts. Their activity in the Fischer-Tropsch synthesis increased to a maximum value as the promoter concentration was increased to about 1 percent, but larger amounts of promoter usually did not improve the activity further. The products from catalysts containing different structural promoters were essentially the same.

Methods have been developed for preparing nitrided-steel turnings. One such catalyst was very active, operated at only 252° C., and yielded products typical of nitrided catalysts (relatively low average molecular weight, containing a high concentration of alcohols). After 6 weeks of synthesis the nitrided catalyst was reduced in hydrogen, and the products from this reduced catalyst were typical of synthesis on a reduced-iron catalyst. Similar results were obtained with nitrided turnings that had been treated with nitric acid after the initial oxidation; however, the activity was considerably lower, requiring an operating temperature of 284° C.

Oxidation of iron, principally by water vapor, probably occurs chiefly inside the pores of reduced, fused catalysts. To test this hypothesis 1/16-inch steel spheres were oxidized 20 percent to provide a layer of active material on the periphery. After addition of alkali and reduction the spheres were tested in the synthesis and removed from the reactor, and the activated surface material was removed by shaking. Its analysis indicated that the surface of the steel spheres had oxidized considerably more slowly than entire particles of reduced, fused-iron catalyst. However, the carbon content of the surface of the spheres was much greater than that of fused catalyst. This experiment is consistent with the postulate that reduced, fused catalysts oxidize predominantly from the inside. The data explain why the activity of fused iron remains essentially constant while the oxide content increases from 0 to 80 or 90 percent.

Sulfur Poisoning of Iron Catalysts

Few data are available on the sulfur poisoning of iron catalysts. As sulfur poisoning is an important factor in practical application of the Fischer-Tropsch synthesis, a systematic study of this problem is now in progress. In one approach catalysts are prepoisoned from liquid-phase contact with known amounts of sulfur compounds dissolved in heptane; in another, the poison is introduced in the vapor

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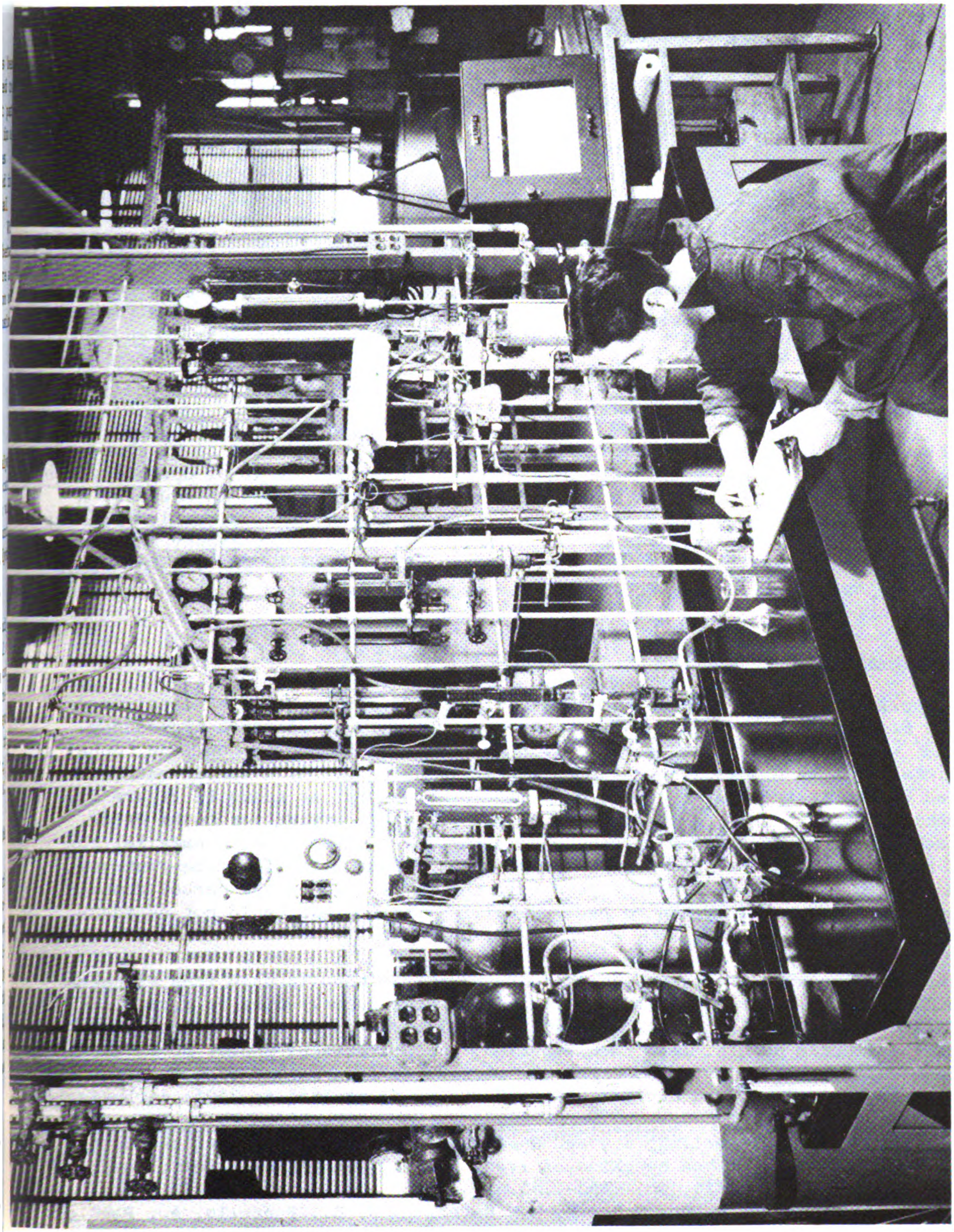

Figure 20. - Slurry studies apparatus.

phase during the synthesis by addition of known amounts of sulfur compounds to the synthesis gas. The rate and extent of decline in activity are the primary means of measuring the poisoning effect. It is expected that information will thus be obtained on the nature of the poisoning process, the relative resistance of iron phases to poisoning, and methods of reactivating catalysts.

In experiments to date with liquid-phase poisoning, reduced fused iron has been prepoisoned by treatment with heptane containing up to 20 mg. of hydrogen sulfide per gram of iron treated. The activity was decreased to about one-half by addition of as little as 4 mg. of sulfur per gram of iron. In experiments with synthesis gas containing hydrogen sulfide (28 gr. of hydrogen sulfide per 1,000 cu. ft. of synthesis gas) the activity decreased to virtually zero in 175 hours, although only 0.9 mg. of sulfur had been introduced per gram of iron. Poisoning during synthesis is therefore probably confined to the outer, active portion of catalyst particles. In prepoisoning, particles probably are poisoned more uniformly, and the effect on activity is less severe.

Chain Branching in Synthesis Products

Few data are available on the extent of carbon-chain branching of Fischer-Tropsch products, and no information is available on the influence of operating conditions on the relative production of straight- and branched-chain molecules. A method has been developed for removing samples of C_4 hydrocarbons directly from catalyst testing units and analyzing by a combination of gas chromatography and mass spectrometry. The combined data make possible a rather complete analysis of components. The method will serve to predict the degree of branching in the higher carbon number fractions.

Carbon Deposition and Carbides

Deposition of carbon is an important factor in disintegration of catalyst during the Fischer-Tropsch synthesis at high temperatures. An electron-microscopy study[75/] was made of carbon deposits produced from carbon monoxide on iron, cobalt, and nickel. These deposits are filaments containing nuclei of a denser material located either in the center or the ends of the filaments, as shown in figure 21. The nuclei appear to be dense particles of metal, metal carbide, or metal oxide. The iron nuclei are metal and carbide disks, the short axis of which parallel the fiber axis of the carbon deposit. The cobalt nuclei are metallic, face-centered, cubic octahedra, in which one axis parallels the fiber axis of the carbon deposit. The nuclei in the nickel deposits had crystalline facets that could not be identified positively.

Hägg iron carbide is the principal carbide phase found in used iron Fischer-Tropsch catalysts, therefore, its physical properties were studied in detail.[76/] Modifications of Hägg iron carbide, exhibiting differences in X-ray-diffraction patterns and Curie points, have been observed; the differences appear to be linked to the mode of preparation. Carburization of reduced iron with carbon monoxide leads to "normal" Hägg iron carbide with a Curie point at 245° to 250°; carburization

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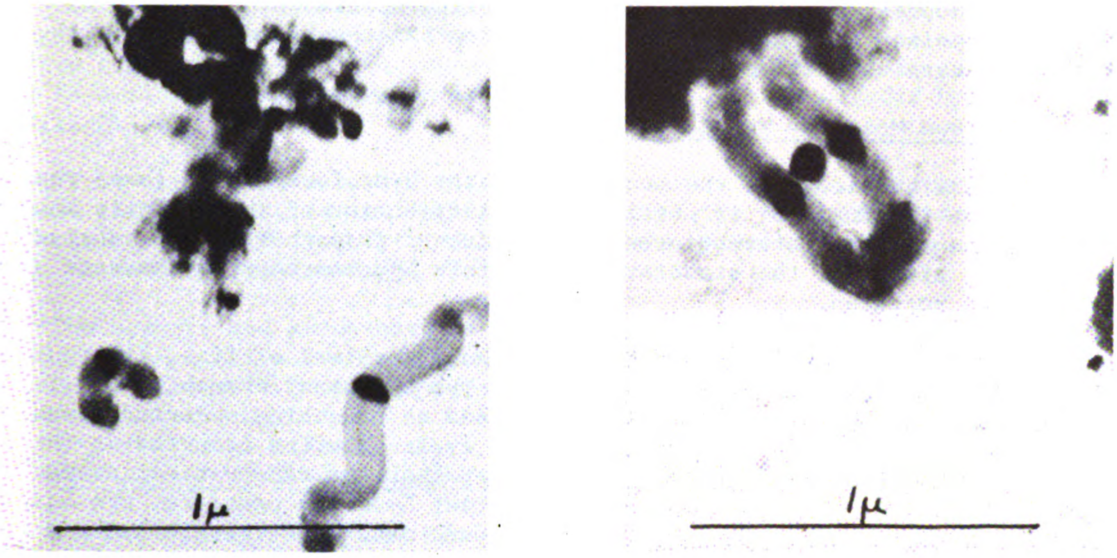
[75/] Hofer, L. J. E., Sterling, E., and McCartney, J. T., Structure of the Carbon Deposited From Carbon Monoxide of Iron, Cobalt, and Nickel: Jour. Phys. Chem., vol. 59, 1955, pp. 1153-1155.

[76/] Cohn, E. M., Bean, E. H., Mentser, M., Hofer, L. J. E., Pontello, A., Peebles, W. C., and Jack, K. H., The Carburization of Iron Oxide With Carbon Monoxide; Modifications of Hägg Iron Carbide: Jour. Appl. Chem., vol. 5, 1955, pp. 418-425.

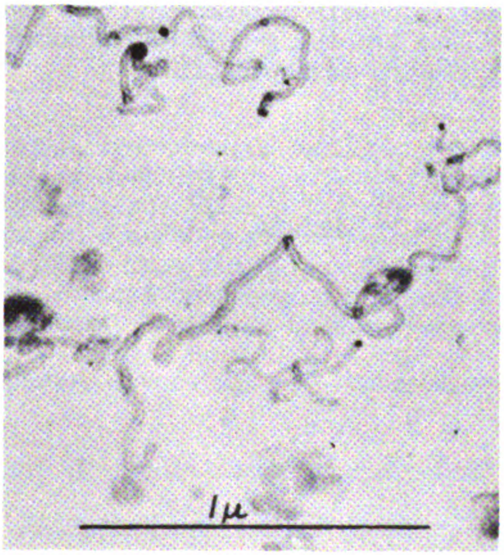


a

Note filaments extending in two directions from central nucleus.

b

Note filaments extending in two directions from central octahedra.



c

Note tubular form of filaments.

Figure 21. - Carbon deposited from carbon monoxide on (a) iron, (b) cobalt, and (c) nickel.

of iron oxide produces Hägg iron carbide with a higher Curie point - up to 276°. The "high-Curie-point" carbide progressively approaches the "normal" form with increasing temperature and time of carburization.

Chemistry of Carbon Monoxide and Carbonyls

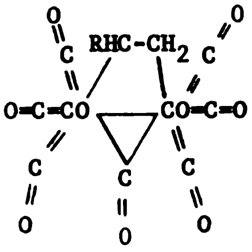
To extend the knowledge of the mechanism of the hydroformylation (oxo) reaction and to relate the information, if possible, to distribution of the products and to the nature of the surface intermediates in the Fischer-Tropsch reaction, a study has been made of the effect of the structure of olefinic hydrocarbons on the rate of the hydroformylation reaction.

The rate decreased in the order, straight-chain terminal olefins, straight-chain internal olefins, branched-chain olefins. The effect of branching was largest at the carbon atom of the double bond and decreased as branching occurred farther in the chain. A minimum in the rate of reaction of cyclic olefins occurred at the six-membered ring. These results, as well as the fact that the rate varies inversely with increasing carbon monoxide pressure at constant hydrogen pressure, are only interpretable on the assumption that a complex between dicobalt octacarbonyl and the olefin is formed in the first step of the reaction; steric hindrance in the formation of this complex is important.

The complex is probably formed in the following manner:

$$Co_2(CO)_8 + RCH{=}CH_2 \rightleftharpoons Co_2(CO)_7.RCH{=}CH_2 + CO$$

By analogy with the acetylenic dicobalt hexacarbonyl and other complexes[77] first isolated in this laboratory, the complex formed by the reaction of olefins and the carbonyl probably has the following structure:[78]



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[77] Wender, I., Friedel, R. A., Markby, R., and Sternberg, H. W., A Bridged Iron Complex Derived From Acetylene and Iron Hydrocarbonyl: Jour. Am. Chem. Soc., vol. 77, 1955, pp. 4946-4947.

[78] Friedel, R. A., Wender, I., Shufler, S. L., and Sternberg, H. W., Spectra and Structure of Cobalt Carbonyls: Jour. Am. Che. Soc., vol. 77, 1955, pp. 3951-3958.

The rate of the reaction did not vary greatly with different solvents; hence the reaction probably does not involve ionic intermediates. The isomerization of olefins that takes place concurrently with addition of synthesis gas to the double bond is a slow reaction that occurs to only a small extent under hydroformylation conditions.

If an olefin has a methyl branch at the double bond, the formyl group adds to the end carbon almost exclusively; thus the reaction of isobutylene with synthesis gas results in approximately a 95-percent yield of isoamyl alcohol and a 5-percent yield of neopentyl alcohol.79/

The spectra and structures of cobalt hydrocarbonyl, dicobalt octacarbonyl, cobalt carbonyl anion, and tetracobalt dodecacarbonyl have been studied. Chromium hexacarbonyl, iron hydrocarbonyl, and iron tetracarbonyl are also being studied by infrared spectroscopy.

Characterization of Products From the Fischer-Tropsch Synthesis

Products from the Fischer-Tropsch synthesis are a complex mixture of hydrocarbons and oxygenated molecules. Special analytical methods have been developed for characterizing these mixtures.

In the mass spectrometer, alcohols produce peaks involving dehydration. Their spectra therefore resemble those of olefins, so that analysis of oil-soluble alcohols in the presence of hydrocarbons is extremely difficult. A simple method has been devised to convert the alcohols into trimethylsilyl derivatives, spectra of which are no longer interfered with by hydrocarbons. Many complex mixtures of Fischer-Tropsch alcohols have been analyzed in the presence of hydrocarbons and other constituents. This method has also been extended to analysis of higher alcohols.

A high-temperature mass spectrometer has been put into operation to obtain mass spectra of compounds boiling over 200° C., which are above the range of the usual room temperature mass spectrometer. The primary purpose of the instrument is to determine a molecular weight distribution of the major classes of compounds found in high-boiling oils and waxes from synthesis processes; however, some information on the structure of these compounds will also be obtained. Spectra of calibration compounds have been determined, including hydrocarbons up to C_{32}, dotriacontane.

A series of 31 aliphatic esters has been investigated, and their mass spectra have been correlated.

SYNTHESIS OF LIQUID FUELS BY HYDROGENATION OF COAL

Pilot-Plant and Bench-Scale Studies

High-Temperature Hydrogenation of Coal

The objective of high-temperature coal-hydrogenation studies was to determine the feasibility of converting coal into refined gasoline in a single step. Autoclave experiments at elevated temperatures (500° to 525° C.) had indicated a shift in product distribution toward low-weight oils and hydrocarbon gases, suggesting the

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79/ Wender, I., Feldman, J., Metlin, S., Gwynn, B. H., and Orchin, M., Formation of Neopentyl Alcohol From Isobutylene in the Hydroformylation Reaction: Jour. Am. Chem. Soc., vol. 77, 1955, pp. 5760-5761.

possibility of converting coal into gasoline in a single step, provided high enough temperatures and appropriate throughputs could be selected without incurring coking. High concentrations of catalyst were believed to favor hydrogenation over coking and gasification. In addition, high temperature, active catalyst, and proper throughput might yield a lower boiling "equilibrium" recycle oil and thus reduce the loss of oil during removal of solids from recycle pasting oil.

The initial bench-scale work was performed in a 32-foot coil of 1/8-inch-inside-diameter tubing, wound in a helix of 4-inch diameter. Rock Springs, Wyo., coal, impregnated with 1 percent of molybdenum after neutralization with sulfuric acid, was used. Coal paste was prepared by mixing 40 weight-percent of coal with 60 of oil having a distillation range of 250° to 312° C. All tests were made by passing the reactants through the coil without recycling at 535° C. and a pressure of 8,000 p.s.i.g.

The data supported the following hypothesis on the events occurring in the tubular reactor: Slug flow of the coal-oil slurry probably occurred at the inlet of the reactor, followed by vaporization of vehicle oil. The vapors attained a velocity comparable to that of hydrogen and were hydrogenated in the vapor phase to gasoline and higher boiling hydrocarbons. During flashing coal was converted to primary products, such as asphaltenes and heavy oils. These fractions were pushed through the tube and formed a rather thin film on the walls. Here, they were hydrogenated to lighter fractions and were swept out as vapor. A steady-state film could thus be established, the thickness of which should be a function of paste rate; hence, in this unit virtually no control of contact time of product oil with hydrogen could be achieved.

The major emphasis in pilot-plant development of high-temperature hydrogenation has been upon removal of the heat of reaction. Several systems, both employing internal hydrogen cooling, have been explored. The first system utilized a single vessel for both the preheating and reaction phases. This design was inadequate, in that only a very small portion of the reactor could be maintained at specified temperatures without incurring excessive temperatures in the preheating zone. The second system employed an external preheater in conjunction with an insulated reactor. After considerable development of instrumentation and operational techniques it has been possible to evaluate the high-temperature, coal-hydrogenation process on a once-through basis.

Initial tests conducted at 8,000 p.s.i.g. with Rock Springs, Wyo., coal using a topped, high-temperature-tar vehicle, indicated that with 1 percent of molybdenum (as ammonium molybdate) temperature control was not possible at and above 480° C. Later studies were conducted with 0.01 percent of molybdenum. Temperature control was comparatively trouble free at 480°, 500°, and 525° C., but formation of "coke" could not be prevented. The conversion of coal to liquid and gases fell from 93 percent at 480° C. to about 88 percent at the 2 higher temperatures.

The gasoline yield, based on feed, was 3 to 3.5 percent. The asphaltene content of the total product oil was 13 percent at 480° C. and 17 percent at 500° C. but only 7.2 percent at 525° C. Hydrocarbon-gas production, based on feed, increased from 14 percent at 480° to 23.4 percent at 500° C. and to 28.7 percent at 525° C.

Hydrocarbonization

"Hydrocarbonization" is a term applied to the carbonization of coal in hydrogen at moderate pressures. The oil can be used for certain fuel applications or refined to gasoline. The hydrocarbon gases are a premium fuel, and the char could be used for power generation or hydrogen production.

Initial experiments were made in a static system at 500 to 2,000 p.s.i. with about 100 grams of 100 to 200-mesh Rock Springs coal. The coal was slowly heated to final temperatures of 500° - 600° C. Extrapolation to pilot-plant scale was unsuccessful due to agglomeration of particles in the reactor bed.

To study the effect of temperature, particle size, and gas velocity on agglomeration, a bench-scale unit was designed to simulate the heating rate employed in a continuous unit. This apparatus, shown in figure 22, consisted of a 2-inch-inside-diameter reactor 3 feet long, provided with a pressurized coal hopper to permit instantaneous coal injection. Preheated hydrogen was fed continuously into the bottom of the coal bed at a velocity of 0.06 ft. per sec. Experiments showed that agglomeration depended on the rate of heating: With slow heating, coal was hydrocarbonized at temperatures as high as 700° C. without agglomeration. With rapid heating mild agglomeration began at 500° C., and severe agglomeration occurred at 600° C. Agglomeration tendencies increased with increase in particle size. The nonagglomerated char produced at 450° C. could be hydrocarbonized at 500° C.; reprocessing this char at 600° and finally at 700° C. also resulted in no agglomeration. This result suggests that coal can be hydrocarbonized continuously by stepwise heating. Further tests indicated that some of these steps could be eliminated. Thermal pretreatment in nitrogen at 400° C. has given similar results. Yields were as follows:

	Yields, weight-percent of moisture- and ash-free coal		
Temperature, °C.	Organic benzene insolubles	Benzene-soluble oil	Hydrocarbon gas
400	85	3	0.1
450	73	11	1
500	62	15	6
600	46	20	15
700	33	20	22

An ASTM-type distillation on the benzene-soluble oil showed that approximately 5 percent distilled at 90° C., 30 percent at 200° C., and 80 percent at 325° C. Above 325° C. thermal cracking occurred.

Hydrocarbonization of a Texas lignite was feasible at temperatures from 400° to 600° C. without agglomeration, regardless of particle size (4- to 300-mesh) and rate of heating.

Laboratory Investigations of Coal and Coal Hydrogenation

Coal Hydrogenation in Autoclaves

The relative activity of impregnated ferrous sulfate and impregnated ferrous chloride in the hydrogenation of Rock Springs, Wyo., coal was studied. The experiments were conducted in horizontal, rotating autoclaves. Under the experimental conditions (3,000 to 8,000 p.s.i.g., 450° and 500° C.) 1 percent of ferrous sulfate was a better catalyst for liquefaction than ferrous chloride. At concentrations of

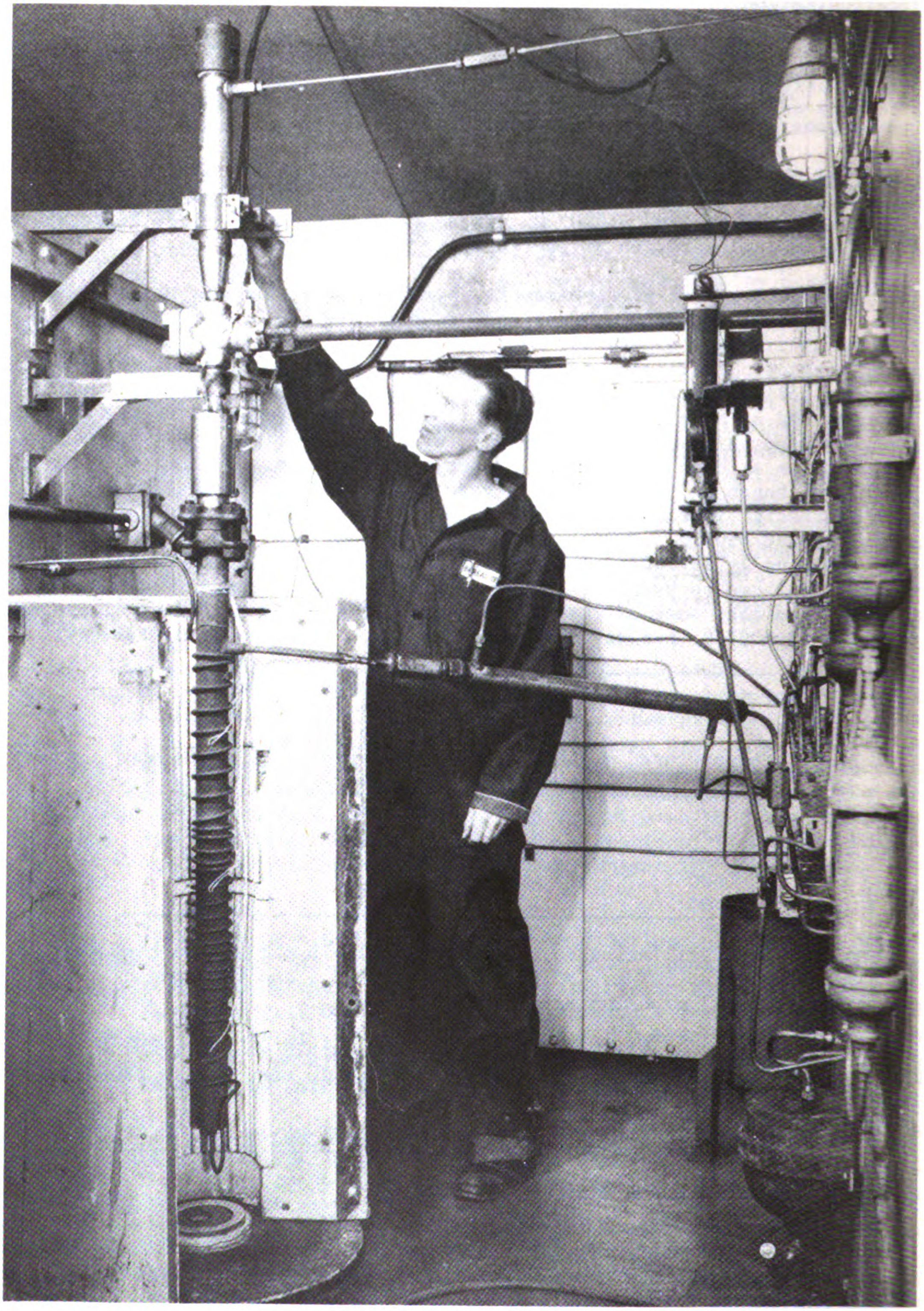

Figure 22. - Bench-scale hydrocarbonization unit.

0.1 percent, both sulfate and chloride were poor catalysts. In the range from 3,000 to 8,000 p.s.i.g. at 450° C., the pressure dependence of activity was quite different for the two catalysts. This suggests that the relative activity at a particular pressure cannot always be predicted from knowledge of the relative activity at a different pressure.

Various metallic naphthenates, with and without activated carbon, were tested as catalysts for the hydrogenation and refining of a coal-hydrogenation middle oil at short exposure times, initial pressure of 3,000 p.s.i.g. (cold), and temperatures up to 525° C. Metallic naphthenate combined with activated carbon proved to be an efficient catalyst.

Microbial Degradation of Carbonaceous Matter

In the recently established microbiological laboratory, bacteriostatic and fungistatic materials in coal have been found to be stable to heat and extractable by boiling methanol or ethanol. A quantitative assay of the potency of the material has been devised, and indications are that these agents are more powerful than previously reported.

Investigation of the microflora isolated from lignites has shown it to be almost identical with that isolated when a soil inoculum is used with lignite as a source of carbon. The organisms isolated represent species of gram negative bacteria, molds of the genus *Penicillium*, and *Actinomycetes*. The studies lend credence to the belief that this microflora is capable of utilizing lignitic material for growth.

Studies have been made on the microbial attack of pure compounds whose structure may be similar to that of portions of the coal structure. Phenanthrene is rapidly attacked by an organism identified as *Pseudomonas fluoresceus*, isolated in this laboratory. Isolation and identification of 1-hydroxy-2-naphthoic acid as an intermediate in the microbial utilization of phenanthrene indicate that this molecule is split asymmetrically by attack at an end ring. Certain organisms survive numerous transfers on mediums containing violanthrone as the sole source of carbon and energy; this indicates that the organisms can utilize the compound, probably the largest system of condensed benzenoid rings ever reported as being attacked.

Spectroscopic Studies of Coal

The infrared spectra of coal and related substances have been determined.[80] Similarities and differences have been found among the infrared spectra of various coals, coal chars, carbohydrate chars, coal and petroleum asphalts, and other materials. The infrared spectrum of Bruceton anthraxylon was investigated from 2 to 25 microns. Specific assignments of spectral bands to molecular structures serve as a basis and limitation for proposed structures of coal. Electron paramagnetic resonance spectra have been determined by the laboratory of Varian Associates. These spectra measure the amounts of free radicals and indicate types and amounts of CH, CH_2, CH_3, and OH groups present in coal and coal products.

Ultraviolet and visible absorption spectra of thin sections of Bruceton anthraxylon have been determined. The absorption is comparatively weak and does not justify the hypothesis that polynuclear condensed aromatics are present in high concentration. Also, structures containing several conjugated double bonds, such as octatrienol, are not likely to occur in coal because the absorption intensities of these molecules are greater than those of most polynuclear aromatics.

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[80] Friedel, R. A., and Queiser, J. A., Infrared Analysis of Bituminous Coals and Other Carbonaceous Materials: Anal. Chem., vol. 28, 1956, pp. 22-30

Rather than attribute the color, the absorption spectra, the reflectivity, and the refractivity of coal to the existence of polynuclear aromatics, free radicals and molecular complexes are considered to be the more logical possibilities. Ultraviolet, visible, infrared, nuclear, and paramagnetic resonance spectra are needed for this study.

Coal Structure by X-Ray Diffraction

Coal may be considered to consist of small, two-dimensional sheets or platelets of condensed six-membered rings. The platelets probably represent the highest molecular weight material in coal and that most refractory to hydrogenation. Some of these platelets are stacked, and some are not. In addition, some coaly material is quite unorganized, so that it must be highly amorphous. Furthermore, some coals contain a liquidlike component whose scattering approximates the scattering from an aliphatic liquid hydrocarbon.

Methods of X-ray-diffraction analysis are being developed to evaluate these various parameters of coal. Provisionally the characteristics of Bruceton anthraxylon have been determined as:

Platelet diameter = 7.0 - 7.6 Å.
Stack height = 7.1 Å.
Organized to unorganized carbon ratio = 70:30.

Sorption Studies on Coal

Sorption data on American coals obtained at the Bureau of Mines agree in essentials with those from foreign laboratories: The sorption of nitrogen at liquid nitrogen temperatures was low on all coal samples examined, and the sorption of methanol and water was very much greater. Helium- and mercury-density measurements of several American coals showed that these coals have sizable pore volumes. Furthermore, the sorption of normal butane and isobutane indicated that a large fraction of the pores has openings of about 5 Å.

Sorption at the lower temperature was limited by a temperature-dependent rate process, probably activated diffusion in pores. At the temperature of liquid nitrogen inert molecules, such as nitrogen and argon, are apparently adsorbed only in larger pores and do not enter the micropore structure. Thus the surface areas computed from these isotherms give the areas of the portions of coal having large pore openings. This value is lower than the "true surface area"; however, it is questionable whether a true surface area can be estimated from gas adsorption on systems of pores of molecular dimensions. On the other hand, sorption of polar molecules appeared to involve weak chemical interactions with polar groups and was accompanied by swelling of the coal substances. These isotherms are characteristic of adsorption on polymeric substances containing oxygen and/or nitrogen.

Optical Activity in Oils Derived From Coal

Several coal-hydrogenation oils and a low-temperature lignite tar have been found to contain optically active constituents. Fractions isolated from these sources give both positive and negative rotations. The optical activity has been found chiefly in the paraffin-naphthenic and hydroaromatic portions of the various neutral oils; little or no activity has been found in the acidic or basic fractions. The oils obtained by treating coal with hydrogen in the presence of added catalyst contain fewer optically active materials than those obtained from low-temperature tar. However, a fraction with high specific activity has been isolated from a Louisiana, Mo., coal-hydrogenation middle oil.

Optical activity has been found in lignite-tar fractions that boil as low as 165° C. Mass spectrometric analysis and examination of the physical properties of this material indicate that the optical activity is due to a C_{10} naphthenic hydrocarbon, which is probably a cyclohexane derivative.

Analytical Methods Applicable to Products of Coal Hydrogenation

Products from coal hydrogenation are a complex mixture of cyclic and straight-chain hydrocarbons and their derivatives. Methods have been developed for identifying phenols by X-ray diffraction and characterizing aromatic mixtures by ultraviolet spectrometry.

X-ray-diffraction analysis is proposed for the positive identification of solid crystalline phenols.[81] If the phenols have a reasonably high melting point, conversion to the phenyl isocyanate derivative for the purpose of making a diffraction pattern is unnecessary. It is also unnecessary to recrystallize the specimen until the melting point is attained. The X-ray patterns of isomers and closely related phenols were found to be distinctly different. Correlations of ultraviolet spectra and chemical structures were useful in analytical work and helpful in understanding the relationship of the structures of ring compounds and their derivatives.

SYNTHETIC LIQUID FUELS PUBLICATIONS

Special problems encountered in developing processes for converting coal to gas, liquid fuels, and coal chemicals were investigated and reported in the technical press.[82 83 84]

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[81] Hofer, L. J. E., and Peebles, W. C., X-Ray Diffraction Patterns of Phenols: Anal. Chem., vol. 27, 1955, pp. 1852-1856.

[82] Ergun, S., Flow Experiments in Studying Kinetics: Ind. Eng. Chem., vol. 47, October 1955, pp. 2075-2080.

[83] Friedel, R. A., and Pelipetz, M. G., Special-Purpose Infrared Cells: Jour. Opt. Soc. America, vol. 45, 1955, p. 822.

[84] Pelipetz, M. G., Salmon, J. R., Bayer, J., and Clark, E. L., Uncatalyzed-Coal Hydrogenolysis: Ind. Eng. Chem., vol. 47, 1955, pp. 2101-2103.

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